WANL-TME-1896

January 1969

Westinghouse Astronuclear Laboratory
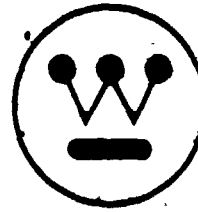
FISSION PRODUCT RELEASE FROM NERVA FUELS

A SUMMARY

(U)

MASTER

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Mr. J. L. Dooling
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Subject: WANL-TME-1896, "Fission Product Release from NERVA Fuels -
          A Summary," (Title Unclassified) dated January 1969.

Dear Mr. Dooling:

        Transmitted herewith for your information are ten (10) copies

of the subject report.

                                Respectfully,

                        ORIGINAL SIGNED BY M. K. WRIGHT

                                M. K. Wright
                                Program Manager
                                NERVA Nuclear Subsystem

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FISSION PRODUCT RELEASE FROM NERVA FUELS

A SUMMARY

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by _J. T. C_   TO BE SEP 12 1973

GROUP 1
Excluded From Automatic Downgrading
And Declassification

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I. INTRODUCTION

(U) The first phase of the Nuclear Engine for Rocket Vehicle Application program (NERVA), is nearing completion. This first phase is concerned with the development of basic technology to support reactor and engine design. As part of this development a series of reactor and engine tests were made at the Nuclear Rocket Development station in Nevada. Attendant to high temperature operation of a gas cooled reactor with unclad fuel elements is the release of radioactive fission products. In order to determine the magnitude of the release of these fission products, a fission product diffusion program was initiated at WANL in 1962.

(U) This report summarizes the experimental methods utilized in the fission product diffusion program over the period September 1962 to September 1968.

(U) The fission product diffusion program consists of two parallel courses:

(U) The first of these is an experimental program conducted in the Physical Sciences Laboratory at WANL. Solid state diffusion studies on the release of fission products from various types of NERVA fuel were made as a function of time and temperature.

(U) The second course of action was the measurement of the quantity of fission products released during the various reactor tests made by WANL at the Nevada Test Site.

(U) Diffusion constants derived from fission product release rates obtained in the laboratory were used as input to a computer code, FIPDIF, which was developed to determine the magnitude of fission product release during NERVA reactor tests. Measurements of the actual fission product release during tests were used to evaluate the accuracy of the calculations. The measured values of fission products released during a test were also used to modify the input to the code. Thus, via an iterative process the accuracy of the predictions was improved.

(U) This report also summarizes the significant results of laboratory experiments on fission product diffusion reported previously and provides a tabulation

of the fission product release and source term measurements made on the NRX
series of reactor tests. This information is used as a basis for calculating
the expected release of fission products from R-series reactors which will be
built for the preliminary flight rating phase of the NERVA Program.

II. FISSION PRODUCT RELEASE DURING NORMAL OPERATION

A. Types of Fuel

(U)   The progressively improved performance of the NERVA reactors, starting
with the KIWI prototypes through the A6, reflects the improvement in the fuel
used in these reactors. The changes in the fuel of the different NRX series
reactors are discussed below in terms of the effect of these changes on fission
product release.

1. Type I Fuel

(C-RD) The KIWI reactors, including the KIWI B-4, were fueled with Type I fuel.
In Type I fuel, an admixture-type fuel, uranium carbide particles, 8 to 10
microns in diameter, are uniformly distributed throughout a graphite matrix.
Some limited release studies have been performed[1] on this fuel which indicate
that the release of fission products was large, as shown in Table I. These data
were not sufficient to derive diffusion constants. The admixture-type fuel was
abandoned for use in the NRX-series reactors because of mechanical problems
arising from appreciable hydrolysis of UC_2 kernels.

2. Type II Fuels

(C-RD) The NRX-A2, A3, A4 and A5 reactors were fueled with a bead-type fuel,
designated as Type II Fuel. In Type II Fuel, the UC_2 is uniformly dispersed
throughout the graphite matrix in the form of microspheres, or beads, with the
average diameter of 100 microns. Around the UC_2 beads, a single 25 micron thick
layer of laminar pyrolytic graphite was deposited in one operation.
(C-RD) So called duplex fuel has been found[2] to have superior UC_2 migration
resistance compared with Type II fuels. The duplex fuel used in release
experiments had two 12.5 micron thick layers of pyrolytic graphite deposited
in two separate operations with a cooling period between operations. This
deposition process results in a similar 25 micron thickness of pyrolytic

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TABLE 1. Fractional release of selected fission products from Type I fuel.

Sample No.	Time (min.)	Temperature (°F)	Sr89	Ba140	Zr95	Y91	Ce/Pr144	Cs137	Mn54
2	30	1600	0.99	0.39	0.00	0.52	0.04	0.04	0.00
3	15	1600	0.96	0.37	0.00	0.42	0.41	0.97	0.00
4	15	1600	0.56	0.57	0.00	0.37	0.06	0.77	0.00
5	5	1800	0.58	0.41	0.00	0.36	0.00	0.78	0.00
6	15	1800	0.78	0.63	0.00	0.37	0.01	0.82	0.00
7	30	1800	0.80	0.80	0.00	0.36	0.08	0.92	0.00
8	60	1800	0.93	0.86	0.00	0.35	0.17	0.95	0.00
9	15	2000	0.99	0.73	0.00	0.52	0.62	0.99	0.00
10	60	2000	1.00	1.00	0.00	0.67	0.99	1.00	0.00

Sample 4 was run in helium at atmospheric pressure.

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graphite surrounding the fuel bead but exposes the UC_2 to lower temperatures during the second coating process.

(C-RD) The slight differences in fabrication methods of Type II and Type II Duplex Fuel do not have any effect on the release of fission products since the major diffusion barrier is a 25 micron thick layer of laminar pyrolytic graphite in either fuel type.

(U) The diffusion characteristics of the major contributors to the total fission product activity in irradiated Type II Duplex Fuel have been carefully determined.[3] The release of all nuclides by diffusion can be described by an equation of the form

$$f = g_1 \exp(-D_1 t) + g_2 \exp(-D_2 t) \tag{1}$$

where f is the fraction of a given nuclide retained in the fuel, t the heating time, c_i the fraction of that nuclide which diffused at a fast rate, characterized by the rate constant D_1, and g_2 the fraction which diffused at a slow rate, characterized by the diffusion constant D_2. The temperature dependence of the rate constants is believed to follow an expression of the form

$$D_1 = D_o \exp(-E/RT_1) \tag{2}$$

where D_1 is the diffusion rate at an absolute temperature T_1, D_o is a frequency factor and E/R an activation energy.

(U) It should be emphasized that equation (1) refers to a given nuclide, for example Sr^{89}. In order to incorporate the two-term rate equation (1) in the FIPDIF Code, artificial '0-isomers' were postulated such that the release of Sr^{89} by diffusion first proceeds rapidly by releasing the Sr-0 isomer, then by the slow diffusion of Sr^{89}. In cases where the release by diffusion proceeds in a one-step process, the term $g_1 \exp(-D_1 t)$ in equation (1) becomes zero and g_2 assumes the value 1.00. The diffusion constants measured in release experiments for selected nuclides from Type II Fuel are summarized in Table II.

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TABLE II. Diffusion constants of selected fission products from Type II fuel.

Elements	D_0 (Sec^{-1})	$1/R$ ($^\circ K \times 10^3$)	$P_1^{(1)}$	$P_2^{(2)}$
Sr, Sn, Ag, Cd, Zn, In	3.98×10^7	33.2		0.85
Sr-O, Sn-O, Ag-O, Cd-O, Zn-O, In-O	8.70	15.5	0.15	
Ba	1.83×10^{-3}	6.0		0.85
Ba-O	9.17×10^{-3}	2.5	0.15	
Ce, Y, La, Kr, Xe, Sb, Pr	7.97	26.0		1.00
I, Cs, Br, Te, Rb	5.38×10^{10}	87.2		1.00
Mo, Zr, Ru, Nb, Tc	0	0		1.00

(1) P_1 is the fraction of concentration of nuclide which diffuses at fast rate (see Equation (1)).

(2) P_2 is the fraction of concentration of nuclide which diffuses at slow rate (see Equation (1)).

3. Type III Fuels

a. Type III Fuel

(C-RD)   In order to predict the fis_ion product release during a hypothetical
loss-of-coolant accident, diffusion data were collected on Type II Duplex Fuel
which had been heated for 30 minutes at 2500 °C before irradiation. The initial
heat treatment was intended to simulate the thermal transient a NERVA reactor
would undergo should a loss of coolant occur. The heat treatment caused the UC_2
to migrate resulting in a dispersion of small UC_2 crystallites throughout the
matrix of the fuel element. The fuel was 'degraded' and was designated as
Type III.

(C-RD)   One set of conditions under which Type II Duplex Fuel degraded to
Type III Fuel was determined[*] as heating for 5 minutes at 2250 °C. Both,
Type II Fuel and Type II Duplex Fuel degrade under the same conditions as shown
in Figure 1. Each point plotted represents the fraction of total gamma activity
released from 5 minutes of heating. The break in the fraction released versus
temperature curve at 2250 °C indicates the onset of degradation.

(U)   The diffusion constants measured[3] for selected fission products in
Type III Fuel are summarized in Table III.

b. Type III NbC Fuel

(C-RD)   FIPDIF predictions based on uncoated Type II Duplex Fuel were high
compared with measured release values from reactor tests. It was reasoned that
the only way by which fission products could reach the outside of an operating
NERVA reactor was by diffusion into the NbC coated coolant channels and then
being swept out of the coolant channels by the hydrogen flow. Thus, for FIPDIF
predictions, diffusion data should be used which had been derived from fuel
samples which had been completely coated with NbC. Such samples were prepared
by the vapor phase decomposition of $NbCl_5$ on 1/4-inch Type II fuel sections at
1800 °C. This process resulted in depositing a one-mil layer of NbC on the

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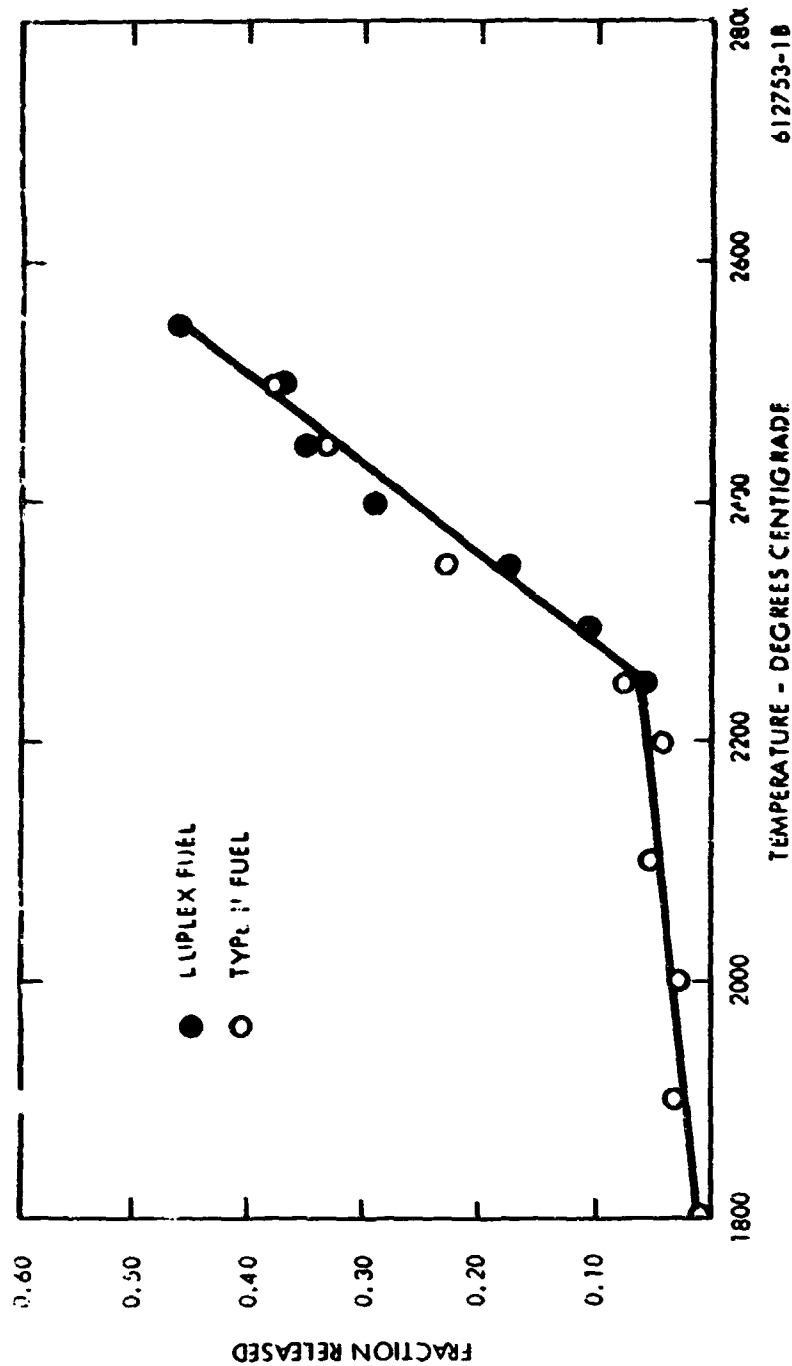
Figure 1. Fractional release of gross gamma fission products from Type II and Duplex Fuels heated for 5 minutes in a vacuum.

TABLE III. Diffusion constants for selected fission products in Type III Fuel.

Element	% Abundance at Zero Time	Representative Nuclide	D_o (Sec^{-1})	E/R ($^\circ K \times 10^3$)
Nb	10.18	Nb^{95}	** 0	0
Sr	8.05	Sr^{89}	1.63	11.4
Rb	7.71	* -	0	0
Y	6.81	Y^{91}	0.379	17.0
Xe	5.98	Xe^{133}	1.63	11.4
Cs	5.86	Cs^{137}	0.477	10.5
Kr	5.73	Kr^{87}	1.63	11.4
Zr	5.62	Zr^{95}	0	0
Br	5.36	* -	0	0
Ba	4.72	Ba^{140}	1800	26.2
Sb	4.70	Sb^{127}	0.779	17.0
I	4.70	I^{131}	2.67	18.2
La	3.70	La^{140}	21.6	25.3
Ce	3.50	Ce^{141,144}	0.779	17.9
Tc	2.97	* -	0	0
Te	2.97	Te^{129}	0.0805	15.1
Mo	2.83	Mo^{99}	0	0
Se	2.37	* -	0	0
Pr	1.65	* -	0	0
Sn	1.45	Sn^{125}	18.4	18.2
Ru	.10	Ru^{103}	0	0
Ag	0.03	Ag^{111}	1	20.0

TOTAL -- 96.99

* Diffusion constants for nuclides of these elements have not been measured.

** Values of zero indicate no diffusion of these elements.

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outside flats and ends of each 1/4-inch fuel section and also added one mil of
NbC coating to the existing one mil NbC linings of the coolant channels. After
coating, the samples were degraded as described above. A summary of diffusion
constants for some fission product nuclides are given in Table IV.[5]

(U)

TABLE IV. Diffusion constants for Type III NbC Fuel.

Nuclide	D_o (Sec^{-1})	E/R(^oK x 10^3)
Sr^{89}	1.35 x 10^{13}	48.1
Sb^{127}	9.27 x 10^{12}	48.1
Ba^{140}	2.29 x 10^9	37.2
La^{140}	7.02 x 10^6	32.1
I^{131}	7.91 x 10^{21}	87.7
Te^{129}	2.61 x 10^{14}	54.0

4. Triplex Fuels

(C-RD)   The NRX-A6 reactor was fueled with an improved beaded fuel, designated
as Triplex Fuel. In this type of fuel the UC_2 kernels, with an average diameter
of 100 microns, were coated with a one micron thick layer of low density, spongy
graphite and with two more layers of istropic pyrolytic carbon to a total coat-
ing thickness of 25 microns. Triplex Fuel was developed to reduce bead damage
due to thermal expansion of the UC_2 kernel during long reactor runs. Diffusion
data were generated on five configurations of Triplex Fuel: Uncoated Triplex
(bores coated), NbC-coated Triplex, completely uncoated Triplex (bores stripped),
NbC-Mo-coated Triplex, and Degraded Triplex.

:. Uncoated Triplex Fuel

(U)   Tr: release of selected fission products from Uncoated Triplex Fuel was
studied on 1/4-inch sections of the 'as received' fuel. The diffusion constants
derived from these release measurements[6] are listed in Table V. A comparison[7]
between the fission product release properties of Type II and Triplex Fuel is
presented in Figure 2.

(U)

TABLE V.   Summary of diffusion constants for metallic fission
           products from uncoated Triplex Fuel.

Nuclide	D_0 (Sec^{-1})	E/R (^{0}K x 10^3)
Sr^{89}	9.66×10^{-3}	30.2
Y^{91}	0.24	21.0
Ag^{111} (a)	1.80×10^{-2}	19.2
Cd^{115}	1.96×10^{-4}	40.6
Sn^{125}	1.30×10^{-4}	42.5
I^{131}	1.53×10^{-9}	73.3
Ba^{140}	1.12×10^{-4}	21.3
La^{140}	1.37×10^{-5}	51.0
Ce^{141} (a)	2.03×10^{-3}	59.2

(a) Hand calculated from three release rate measurements.

(U)   Gross gamma release fractions from samples used to determine the degrada-
tion threshold are given in Table VI. The gross gamma release data for samples
heated for 5 minutes as a function of temperature[7] are shown in Figure 3.

Figure 2. Comparison of fission product release from Triplex and Type II Fuel heated at 2000 oC for 30 minutes.

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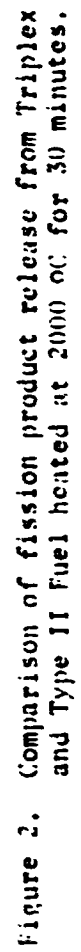
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TABLE VI. Degradation Experiment on Triplex Fuel.

Time Minutes	Temperature ^oC	Gross Gamma Release
5	2100	0.018
5	2200	0.033
5	225C	0.019
5	2300	0.042
5	2350	0.104
5	2400	0.183
5	2450	0.244
5	2500	0.292
5	2300	0.042
10	2300	0.051
15	2300	0.080
20	2300	0.114
22.5	2300	0.091
25	2300	0.100
30	2300	0.144
45	2300	0.263

(C-RD)   It may be seen that at 2350 ^oC, the gross gamma release is much higher than the expected release from an extrapolation of the lower temperature data. It appears that for Triplex Fuel heated for 5 minutes, at temperatures of 2300 ^oC

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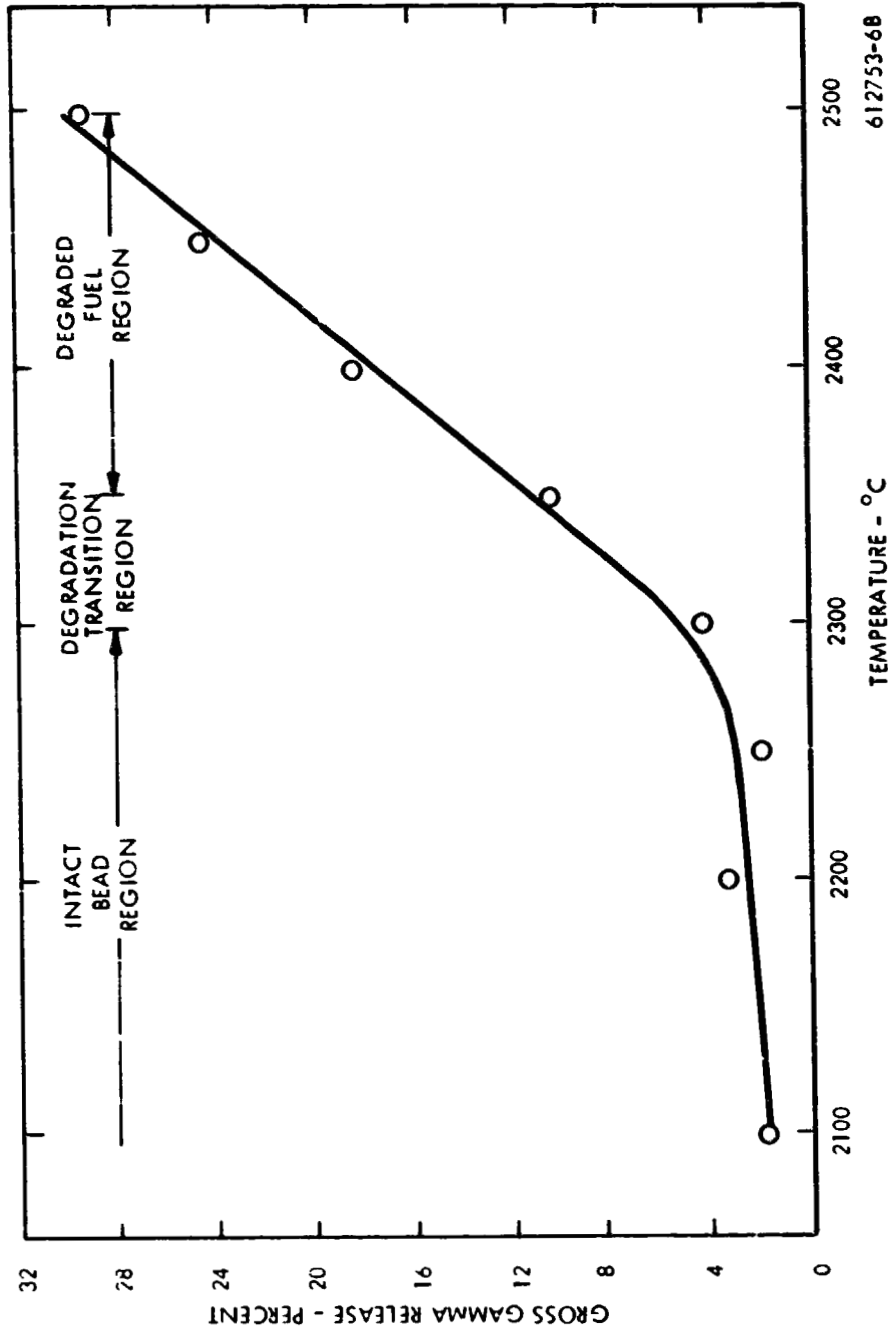
Figure 3. Fission product release from Triplex Fuel as a function of temperature during five-minute heating periods.

and lower, fission product release is relatively slow and infers that the fuel
beads are intact. At temperatures of 2350 ^{O}C and higher, fission product
release is relatively fast. This is a characteristic of degraded fuel beads.
The transition region from intact to degraded fuel is between 2300 ^{O}C and
2350 ^{O}C. A similar experiment[4] on Type II NERVA Fuel showed this transition
region to be somewhat lower, between 2200 and 2300 ^{O}C.

(U)   Although the fuel beads appear intact at 2300 ^{O}C when heated for 5
minutes, it may be seen in Figure 4 that degradation occurs at this temperature
when the fuel is heated for a sufficient time. At some time between 25 and 30
minutes there is an increased rate of gross gamma release indicative of fuel
degradation.

        b. NbC Coated Triplex Fuel

(C-RD)   To derive diffusion constants, release measurements of some fission
product nuclides were carried out on 1/4-inch fuel sections whose outside flats
and ends had been coated with a one mil thick layer of NbC; the coolant channels
were lined with a 2 mil layer of NbC. The diffusion constants derived from these
release measurements[6] are summarized in Table VII. It should be mentioned here
that the diffusion constants listed in Table VII are not necessarily to be used
as input data to the FIPDIF Code for release predictions for NRX-A6 type reactors.
The constants finally accepted for FIPDIF computations have been modified by
postmortem release measurements on the NRX-A6 reactor and are discussed and listed
in Section C of this chapter.

        c. Completely Uncoated Triplex

(C-RD)   The samples used for all preceding experiments were obtained from
production fuel elements which were processed completely including the NbC coat-
ing of the channels. Diffusion constants designated "Uncoated Triplex" were
derived from experiments using this type of fuel. The question arose concerning
the effect of this channel coating on the release of fission products, since
the surface area of the channels accounts for about 52 percent of the total

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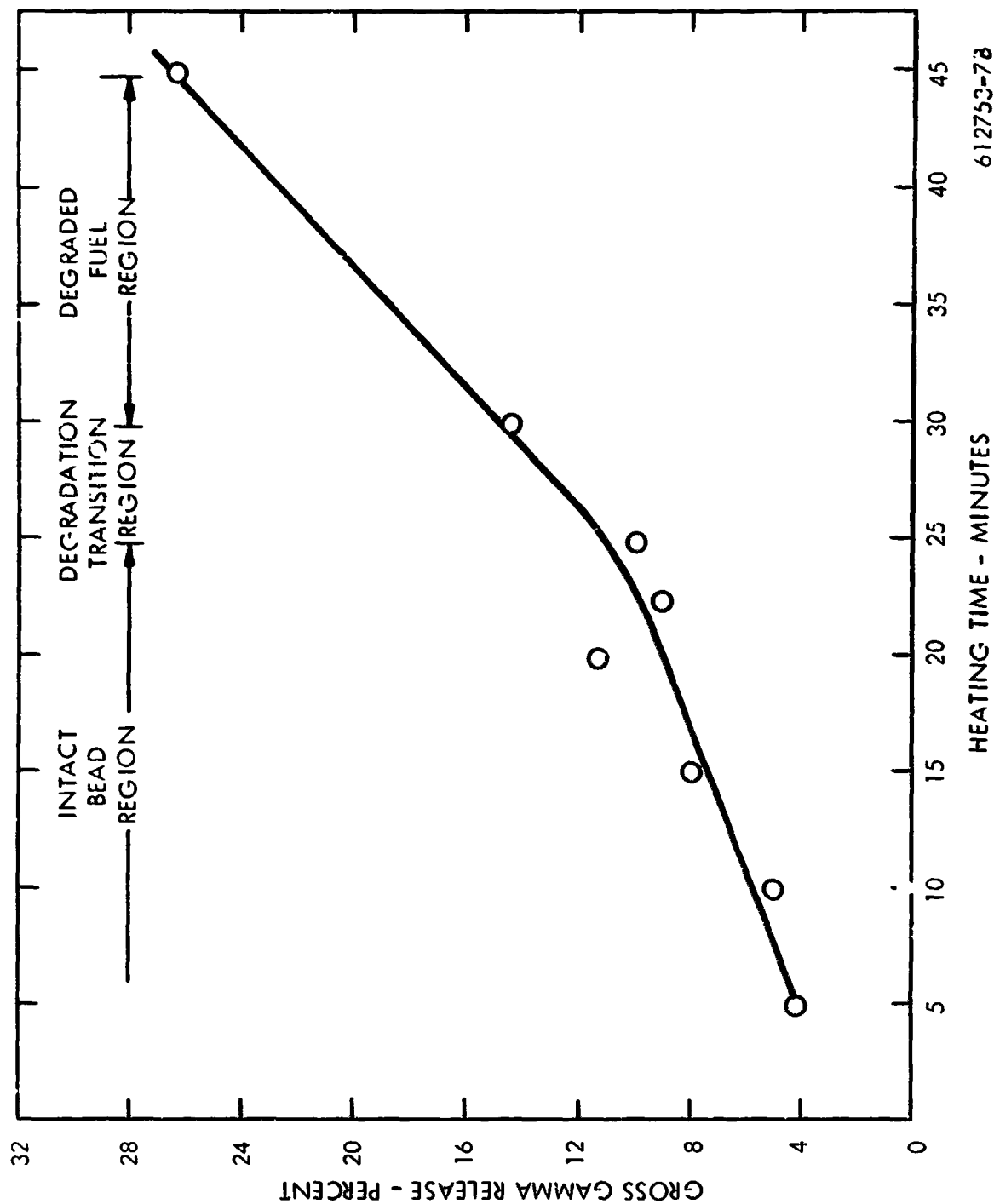
Figure 4. Fission product release from Triplex Fuel as a function of time at 2300 °C.

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HEATING TIME – MINUTES

GROSS GAMMA RELEASE – PERCENT

INTACT BEAD REGION

DEGRADATION TRANSITION REGION

DEGRADED FUEL REGION

(U)

TABLE VII. Summary of diffusion constants for metallic
fission products from Nb oated Triplex Fuel.

Nuclide	D_o (Sec^{-1})	E/P($^{\circ}$K x 10^3)
Sr^{89}	0.29	35.5
Y^{91}	1.94 x 10^{-4}	14.1
Cd^{11}m	2.76 x 10^{-2}	58.2
Sn^{125}	1.95 x 10^{-2}	27.0
Ba^{140}	3.62 x 10^{-2}	29.3
La^{140}	0.43	54.9

geometric surface area of a 1/4-inch slice of fuel. A special fuel element was
obtained which was processed in the same manner as the other elements except for
the final NbC coating of the channels. Eight 1/4-inch slices of fuel from this
element and eight slices from a channel coated element were used in an experiment
to determine the channel coating effect. Two samples of each type were used as
controls and the remaining 12 were subjected to post-irradiation thermal anneals
at 2100 $^{\circ}$C. Two samples of each type were heated for 15 minutes, two for 30
minutes and two for 60 minutes. The samples were heated individually, not
simultaneously. The gross gamma release was determined by gamma counting before
and after each heat treatment. The samples were then dissolved and analyzed
radiochemically for Sr^{89}, Cd^{115m}, Sn^{125}, and Ba^{140} to determine specific nuclide
release. The results obtained are summarized in Table VIII and presented
graphically for the 60 minute heating period in Figure 5. The largest variation
appeared in the gross gamma release. Little significance should be attached to
this, however, because the composition of gross fission product activity changes

with time, and the heating of the samples covered a period of seven days. Other experimental data shows that the gross gamma release decreases with sample age when other conditions remain constant. Since the channel coated samples were heated last, some reduction in gross gamma release was expected. Sample age does not effect the release values for specific nuclides because all samples are corrected to a common time. In general slightly greater release of specific nuclides was observed from the completely uncoated samples. The exception to this was Cd^{115m}, for which the results were reversed. The experimental errors involved could account for the variations in either case. It is concluded, therefore, that within the experimental error limits, fission product release from completely uncoated fuel is the same as for channel coated fuel.

d. Molybdenum-Coated NbC Triplex Fuel

(C-RD) The most recent improvement in fuel development is to deposit an additional layer of molybdenum in the coolant channels. A preliminary experiment to determine the effect of this coating on fission product diffusion was performed and reported previously.[b] Another experiment was conducted using this type of fuel at higher temperatures to acquire more complete data.

(C-RD) This experiment consisted of eighteen 1/4-inch slices of Triplex Fuel. Two slices, which were used as controls, were sectioned from a production run fuel element and used without further processing. The other sixteen samples, also taken from a production run fuel element, were given an additional coating of NbC. This coating, applied by vapor deposition of $NbCl_5$ at 1800 $^{\circ}$C, was 1 mil thick on the surfaces which were previously uncoated and approximately 2 mils thick in the coolant channels. An additional coating of molybdenum was then placed on the NbC surface. This was also done by the vapor deposition method. The average thickness of the molybdenum layer was calculated to be about 0.2 mil. The samples were irradiated to a level of approximately 5×10^{13} fissions each, and then heated individually at varying times and temperatures in vacuum. The heating time periods ranged from 5 minutes to 60 minutes. The selected temperatures were corrected for emissivity[a], required for coated samples, of apparent readings of 2100 $^{\circ}$C, 2270 $^{\circ}$C, 2380 $^{\circ}$C

TABLE VIII. Comparison of the fractional release of selected
fission products from uncoated and channel-coated Triplex
Fuel at 2100 °C.

Nuclide	Fractional Release			Sample Type
	15 min	30 min	60 min	
Sr-89	.520	.510	.648	Uncoated
	.491	.470	.608	Channel Coated
Cd-115m	.187	.276	.552	Uncoated
	.161	.366	.635	Channel Coated
Sn-125	.232	.376	.532	Uncoated
	.098	.218	.506	Channel Coated
Ba-140	.532	.530	.678	Uncoated
	.379	.436	.609	Channel Coated
Gross Gamma	.090	.096	.186	Uncoated
	.059	.064	.130	Channel Coated

and 2500 °C produced true temperature values of 2000 °C, 2100 °C, 2200 °C and
2300 °C, respectively. After heating, the samples were dissolved and analyzed
radiochemically to determine release fractions. The results are presented in
Table IX. These values were compared with values obtained from uncoated fuel
and NbC-coated fuel. In general, for corresponding times and temperatures, the
greatest release came from uncoated fuel, the smallest release was from NbC-
coated fuel, and the NbC-molybdenum-coated fuel values fell in between. An
example of this is given in Figure 6 which shows the release of Ba140 from the
three types of fuel. The isotherms for NbC-coated fuel are not exactly the same
temperatures as the others because of emissivity corrections, but they are close

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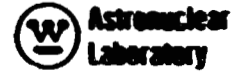
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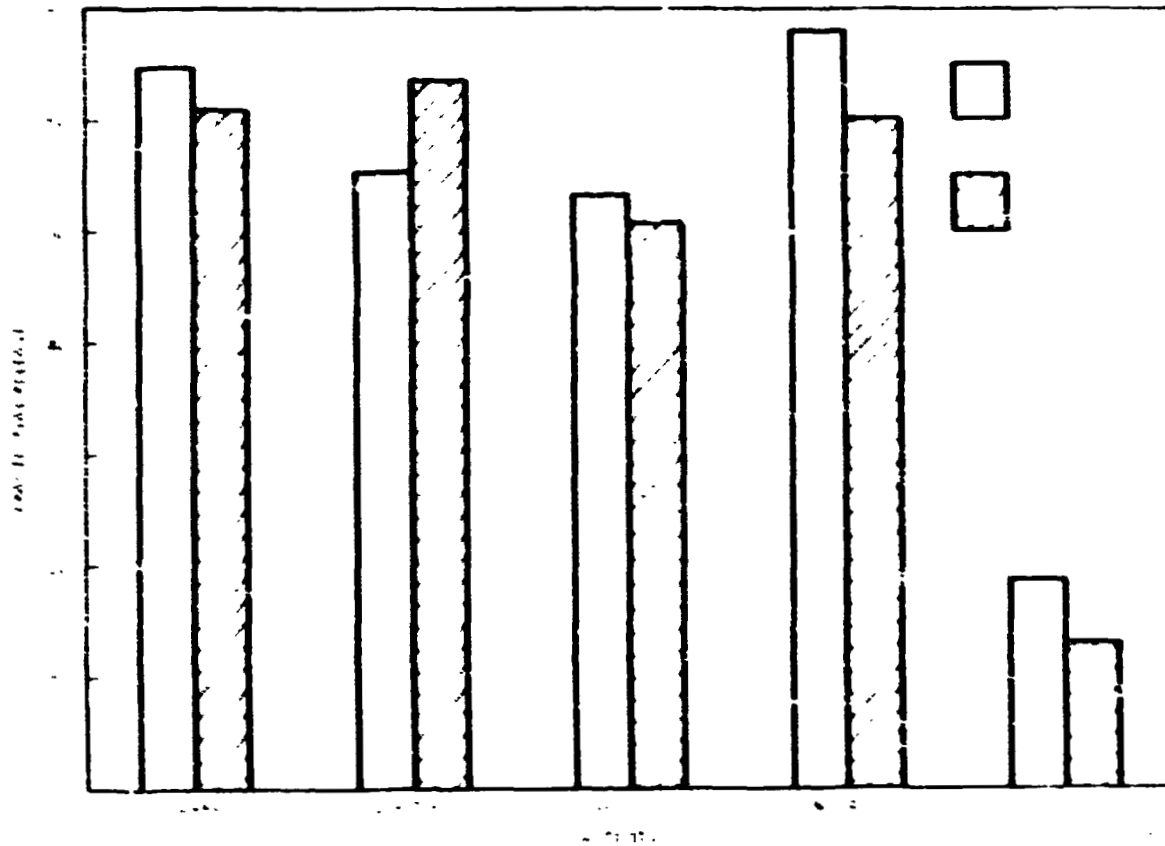
Figure 5. Release of fission products from uncoated and channel-coated Triplex Fuel after heating for 60 minutes at 2100 °C.

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TABLE IV. Fractional release of fission products from hot, oxwhydrogen coated triplex fuel.

Temp. °C	Duration	Xe-133	Y-91	Ce-143e	Ba-125	I-133	Ru-103	La-140	Ce-141	Gross (γ)
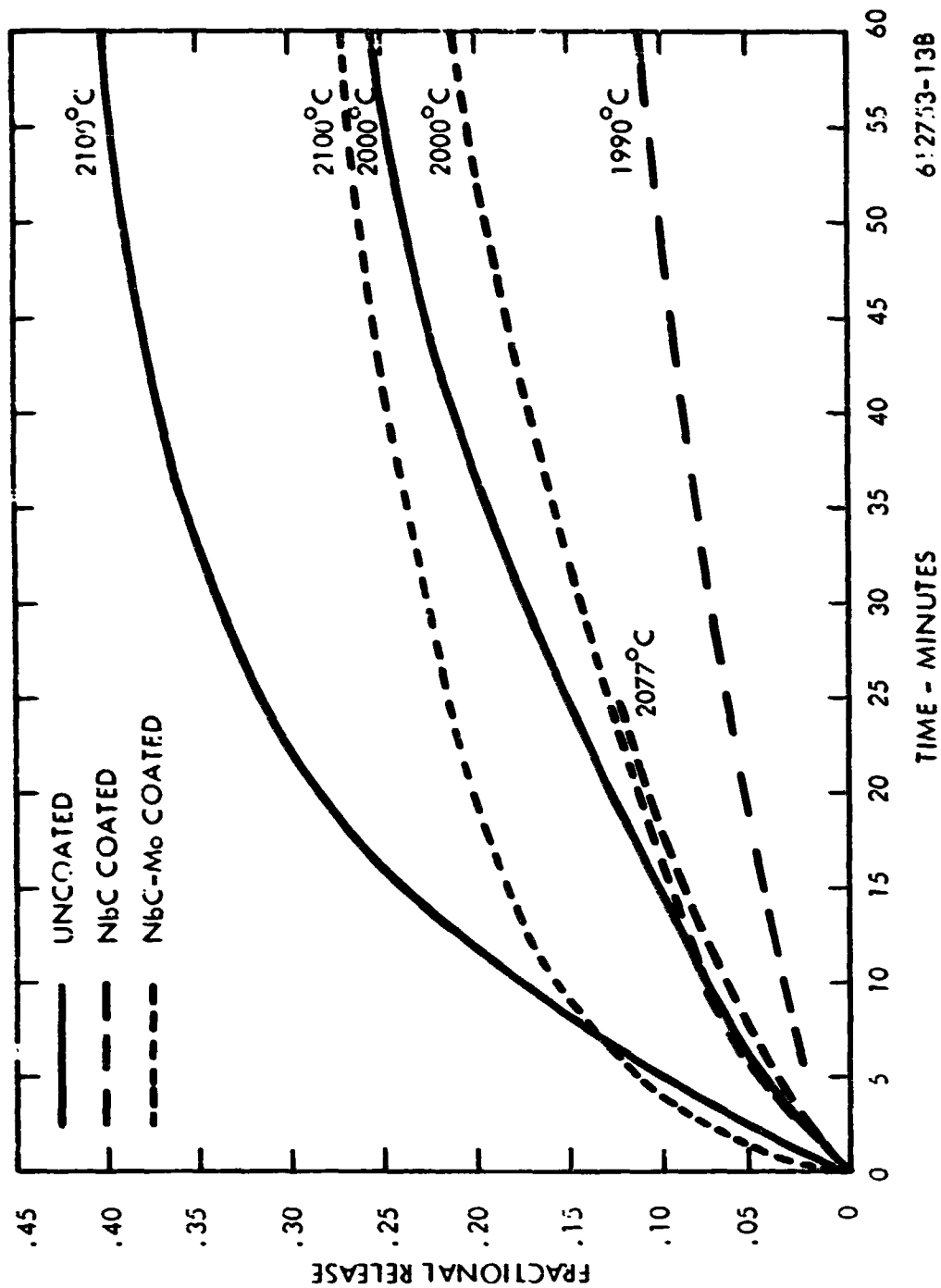
Figure 6. Release of Ba^{140} by diffusion from uncoated, NbC-coated and NbC-Mo-coated Triplex Fuel.
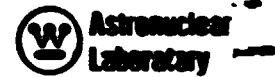
enough for comparison. Since it is doubtful that molybdenum enhances diffusion, the results obtained were probably due to experimental uncertainties. One source of error could be slight clouding of the temperature monitor sight glass while heating the molybdenum-coated samples. Another source might be the temperature correction curves used to convert apparent temperatures to actual temperatures.[8] These correction curves had been derived from emissivity measurements on Mo-NbC-coated samples which had been previously heated once to 2000 °C.

(C-RD) Although the results were not completely satisfactory, it is concluded that a thin coating of molybdenum has little or no effect on fission product release.

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B. Summary of Reactor Runs and Release Data

(U)   From 1964 through 1967, five reactors in the NRX series were tested at various power levels and run times. Each operation of a reactor was designated as an Experimental Plan. The early Experimental Plans for each reactor, usually EP-1, EP-2, and sometimes EP-3, were criticality and calibration measurements. Fission product release from the reactor during these runs was insignificant because activity generation was small and operating temperatures were low. During the later Experimental Plans, however, when activity generation was large and operating temperatures high, fission product release became significant, and was measured whenever possible. Data pertaining to these runs are given in Table X. NRX-A1 was omitted from this tabulation because it was a cold flow test and no activity was generated. The integrated power values given are those which were recorded by the power integrators at the Control Point. The number of fissions was determined by multiplying the megawatt seconds by the conversion factor 3.245×10^{16} fissions per megawatt second. The total integrated power and total fissions for each reactor are included in the table.

(U)   The total release of fission product activity by diffusion was measured for each of the five reactors by post-operational fuel analysis. Release data for these reactors have been reported previously.[3,4,9-10,11]

(U)   A comparison of the release of selected nuclides from the various NRX reactors is presented in Table XI. The release activity is given in terms of curies at time of shutdown. This was calculated by multiplying the fractional release by the total amount generated from all Experimental Plans. For example, during the NRX-A6 test, 1.46×10^{23} fission events occurred. This number of fissions potentially produces 2.966×10^{4} curies of Sr^{89} at zero time. The expression potentially is used because all mass 89 atoms are not Sr^{89} atoms immediately, but become Sr^{89} atoms through decay of mass 89 chain precursors. Post-operational fuel analysis gave a fractional release value of Sr^{89} of 0.0296.

TABLE X. NRX Series Reactor Test Data

Reactor	Experimental Plan	Date	Maximum Power (Megawatts)	Approx. Run Time (Minutes)	Integrated Power (Megawatt-Seconds)	Number of Fissions Produced In Fuel
NRX-A2	EP-4	9-24-64	~1100	6	3.30×10^5	1.07×10^{22}
	EP-5	10-15-64	~60	16	0.43×10^5	0.14×10^{22}
	Total				3.73×10^5	1.21×10^{22}
NRX-A3	EP-4	4-23-65	~1100	4	3.18×10^5	1.03×10^{22}
	EP-5	5-20-65	~1100	13	8.45×10^5	2.74×10^{22}
	EP-6	5-28-65	Mapping	43	4.33×10^5	1.41×10^{22}
	Total				15.96×10^5	5.18×10^{22}
NRX-A4	EP-2B	2-3-66	350	-	3.88×10^5	1.26×10^{22}
	EP-2C	2-11-66	339	-	4.31×10^5	1.40×10^{22}
	EP-3	3-3-66	1175	2 + Mapping	8.69×10^5	2.82×10^{22}
	EP-4	3-16-66	1020	13	9.98×10^5	3.24×10^{22}
	EP-4A	3-25-66	1090	14	9.98×10^5	3.24×10^{22}
	Total				36.84×10^5	11.96×10^{22}
NRX-A5	EP-3	6-8-66	1080	15	1.12×10^6	3.64×10^{22}
	EP-4	6-23-66	1100	15	1.12×10^6	3.64×10^{22}
	Total				2.24×10^6	7.28×10^{22}
NRX-A6	EP-3	12-7-67	290	2	$.03 \times 10^6$	$.01 \times 10^{23}$
	EP-3A	12-15-67	1150	60	4.47×10^6	1.45×10^{23}
	Total				4.50×10^6	1.46×10^{23}

TABLE XI. Activity (Curies) at shutdown released by diffusion from NRX reactors.

Nuclide	NRX-A2	NRX-A3	NRX-A4	NRX-A5	NRX-A6
Sr-89	-	139	451	138	878
Y-91	127	298	-	268	532
Zr-95	49	339	-	42.6	104
Ag-111	-	-	134	116	255
Cd-115m	0.18	1.31	4.34	3.06	6.13
I-131	158	745	1840	<1075	737
Ba-140	173	1078	1985	590	1516
Ce-141	79	-	-	-	963
Ce-144	4.8	-	-	-	80
Gross Gamma	3.4×10^6	3.7×10^6	1.35×10^7	7.08×10^6	4.0×10^6
Gross Gamma*	3.5×10^5	8.0×10^5	3.21×10^6	1.77×10^6	4.0×10^6

*Normalized to 60 minutes after beginning of full power.

Multiplying this value by the curies produced gives 878 curies of Sr^{89} released
from the reactor by diffusion. All values listed for specific nuclides were
calculated in this manner. The gross gamma values were calculated by multiplying
the fractional release by a factor obtained from the curve given in Figure 7
where the decay of gross fission product activity is plotted as a function of
time.[12] Since the factors obtained from this curve differ widely over the
range of reactor run times, a double entry was made in Table XI for gross gamma
release. The first value gives curies released at time of shutdown and the
second value given is normalized to 60 minutes from the beginning of full power
operation. No attempt was made in Table XI to resolve data for multiple runs
or to correct for decay between runs.
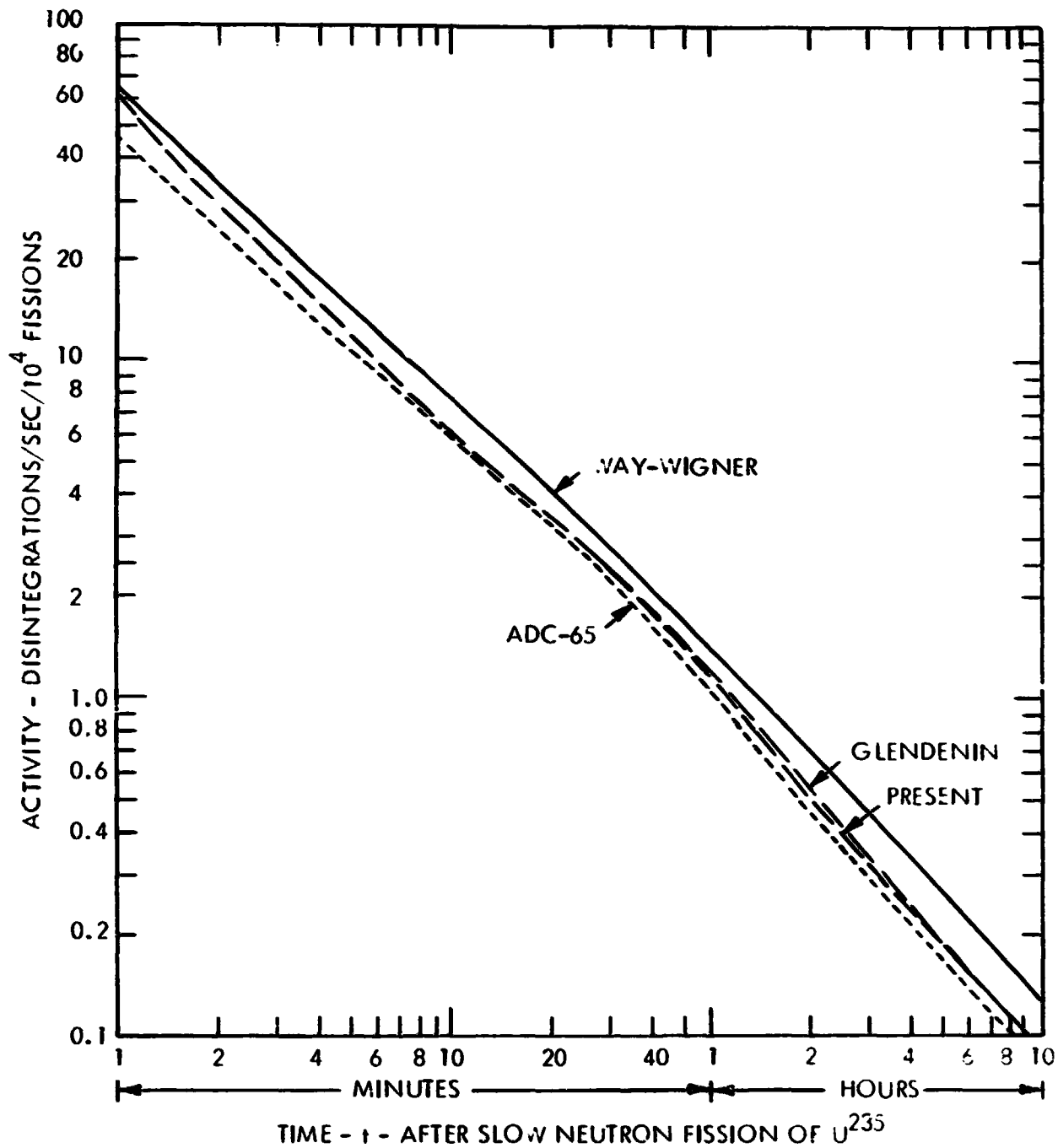
Figure 7. Gross fission product decay as a function of time.

C. Latest Diffusion Constants

(U)   The diffusion constants used to compute the release of fission products
from the A6 reactor are summarized in Table XII. Several comments are in order
here:

(U)   The constants listed in Table XII apply to intact, i.e., non-degraded,
Triplex Fuel.

(U)   All constants, except that for Rb, have been derived both from release
studies in the laboratory and from measured A6 release fractions.

(U)   Fission products whose contribution to the total generated inventory is
less than five percent of the total, such as Zn, Ga, Ge, As, Se, Br, Kr, Nb, Tc,
Rh, Pd, and In, were assigned zero diffusion constants. No attempt had been
made to measure release rates of the above elements in the laboratory. The release
rates for Zr, Mo, Ru, Xe, Te, and Cs, however, had been determined and were found
so small that one had to conclude that these elements do not diffuse measurably in
the 1800 to 2300 ^{0}C temperature range.

(U)   Because of its fast rate of diffusion in the 1800 to 2200 ^{0}C temperature
range, the diffusion constants for Ag were derived from release rate measure-
ments taken at 1200, 1400, and 1600 ^{0}C. These measurements are discussed in some
detail in Section A of Chapter IV.

(U)   The release of mass chain 89 is the sum of the individual release
fractions of Br, Kr, Rb, and Sr isotopes. Bromine and krypton were assigned zero
diffusion constants because of their small contribution to the total fission
inventory. Rubidium was assumed to behave like cesium, i.e., it would not diffuse.
Thus, only the Sr diffusion constant would determine the release of the 89 mass
chain. However, under above assumptions, the prediction of the release of mass
chain 91 was a factor of 5 higher than the A6 measurement. To bring the FIPDIF
prediction more in line with the A6 measurement, it was postulated that Rb
diffused at a finite rate and an artificial Rb diffusion constant was developed.

TABLE XII. Latest NbC Diffusion Constants for Triplex Fuel.

Element	D_o (Sec^{-1})	E/R($^\circ$K x 10^3)
Rb	0.155	18.0
Sr	8.00 x 10^{-2}	22.0
Y	1.94 x 10^{-4}	13.3
Ag	0.760	12.0
Cd	2.76 x 10^{-2}	29.3
Sn	1.94 x 10^{-2}	10.4
Sb	7.00 x 10^8	80.0
I	7.00 x 10^8	82.0
Xe, Te, Cs	0	0
Ba	2.13 x 10^{-2}	17.9
La	.472	23.3
Ce	.472	24.5

III. FISSION PRODUCT RELEASE DURING LOSS OF COOLANT CONDITIONS

A. Summary of Past Experiments

(U) Experiments have been performed to determine the quantity of fission product activity which might be released under accident conditions. In the event of a loss-of-coolant accident, the temperature of the fuel will rise beyond the degradation point. When this occurs, diffusion is much more rapid because the pyrolytic graphite coating on the UC_2 is damaged; this coating is the major diffusion barrier under normal conditions. An example of the high temperature effects on the pyrocarbon coating in Type II fuel can be seen in Figures 8 and 9. To determine the diffusion characteristics of fission product activity under the loss-of-coolant conditions, the following experiments were performed.

1. Simulated Loss-of-Coolant Profile

(C-RD) Analytical studies using the NOFLOW computer program[14] predict temperatures above the fuel degradation threshold within seconds after a loss-of-coolant accident. An experiment was devised to simulate a loss-of-coolant accident and determine the gross fission product release. A 1-1/2 inch section of 19 hole hexagonal Type II fuel was used. This sample was irradiated, allowed to decay for 24 hours then heated in a vacuum (10^{-4} torr) for one hour at temperatures which varied between 2080 $^{\circ}C$ and 2550 $^{\circ}C$ as shown by the temperature profile in Figure 10. This temperature profile (maximum average core temperature) was predicted by the NOFLOW code for a 13-minute full power operation followed by loss of coolant. As a result of this heating procedure, 66.7 percent of the gross gamma activity was lost. A FIPDIF prediction for these conditions, using diffusion constants for Type III fuel, gave a release value of 67.3 percent which agreed very well with the experimental results. These results were indicative of what could be released from the fuel under these conditions. However,

Figure 8. UC$_2$ beads in 'as received' Type II Fuel
(Magnification 250X).
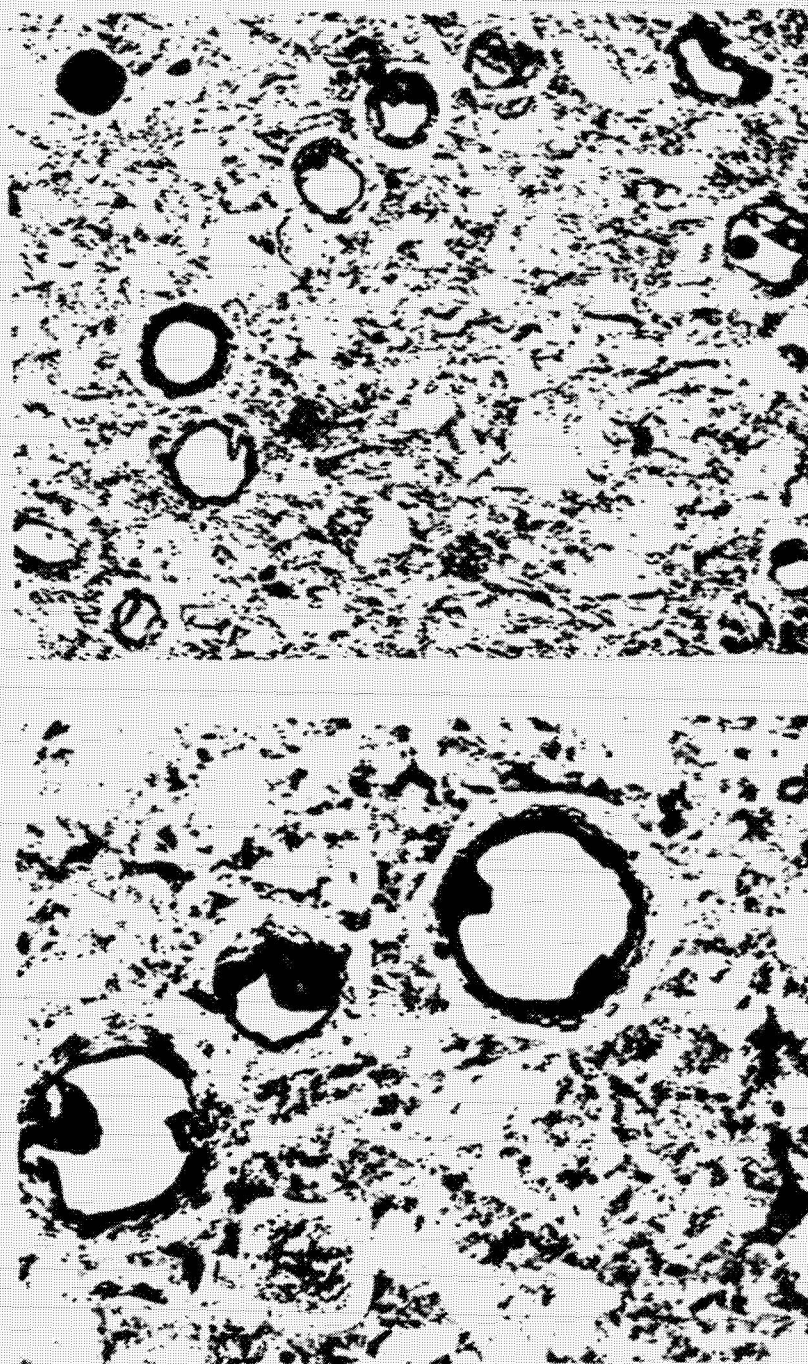
Figure 9. Type II Fuel material heated at 2500 °C for 5 minutes.
(Top 100X, Bottom 200X)

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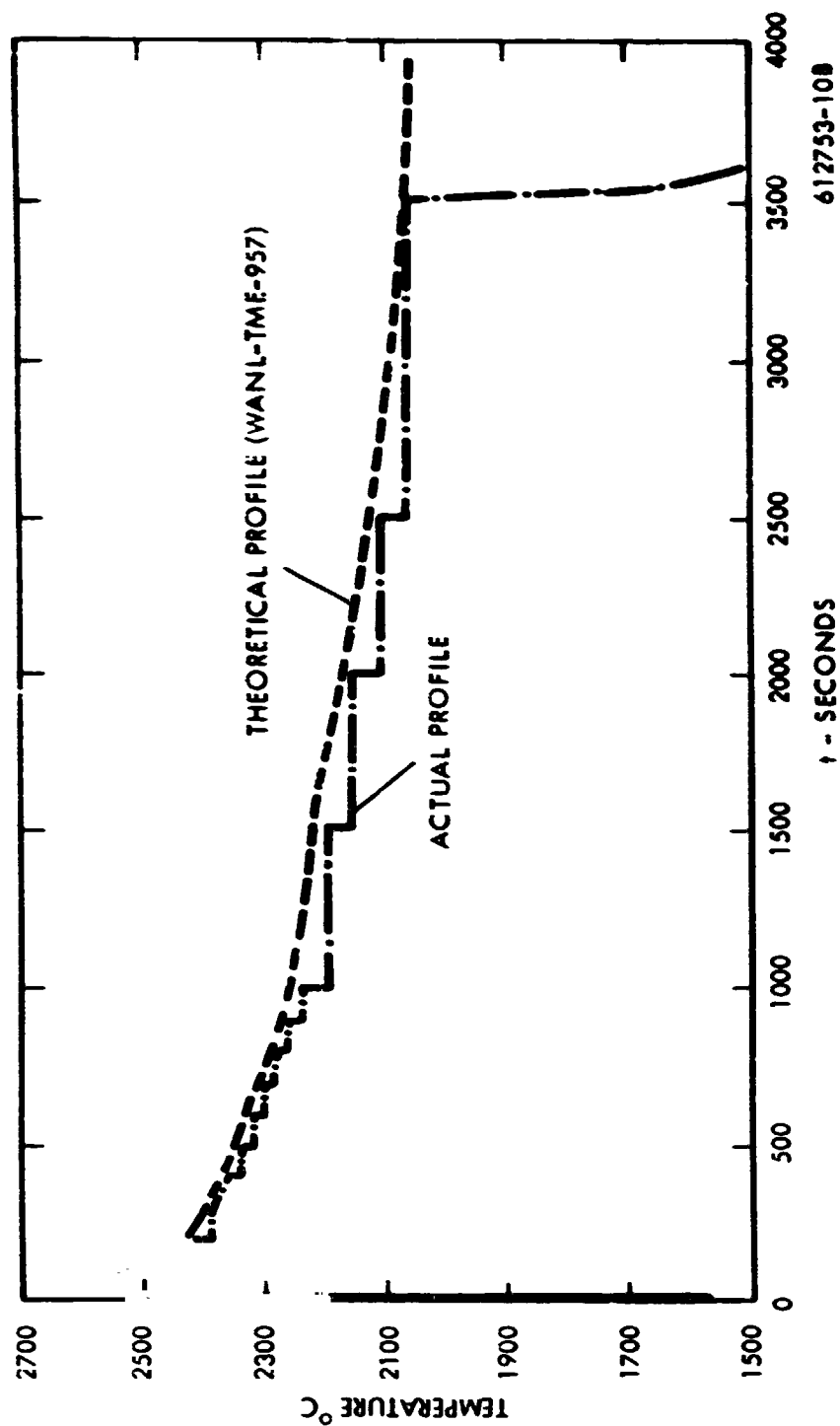
Figure 10. Temperature profile for simulated Loss-of-Coolant accident.

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it does not necessarily mean that this quantity of activity would be released from the reactor proper.

(U) Figure 11 shows four paths for diffusion of fission product activity through NERVA fuel. Paths 1 and 2 pertain to normal operating conditions where the fuel remains intact. The rate limiting step here is the diffusion through the pyrolytic graphite coat. The diffusion paths in 3 and 4 would represent diffusion through degraded fuel where the major diffusion barrier has been destroyed and bulk geometry may become rate limiting. Consequently, although a measure of fission product loss from small laboratory samples is a good measure of the fission product loss from the reactor proper for normal operating conditions, this does not necessarily apply to the loss-of-coolant case. To better understand release from the reactor proper under loss-of-coolant conditions a series of experiments designated "Unit Cells" were conducted.

2. Unit Cells

(C-RD) In order to simulate the reactor components and geometry from a fission product release standpoint, experiments were conducted using individual unit cell assemblies. Each unit cell contained 4 inch long segments of irradiated NERVA fuel, a non-fueled graphite center element, a support plate, orifices, and a modified NERVA tie rod assembly. The geometry of an assembly was chosen to represent a unit cell of the NERVA core and thus provide identical diffusion path lengths for fission product release. During the heating process the unit cell assembly was contained in a hollow graphite cylinder, 3 inches in diameter. The unit cell construction is illustrated schematically in Figure 12 and pictorially in Figure 13. A list of unit cell components is given in Table XIII.

(U) A total of five unit cell experiments were run. The first of these was a feasibility study with non-irradiated fuel to test the heating rates of the vacuum furnace to be used, and to acquaint personnel with the complete experimental operation. The first experiment using irradiated fuel was not successful due to a failure of the unit cell components early in the heating run. With some modification the next three unit cell experiments numbers 2, 3, and 4, were

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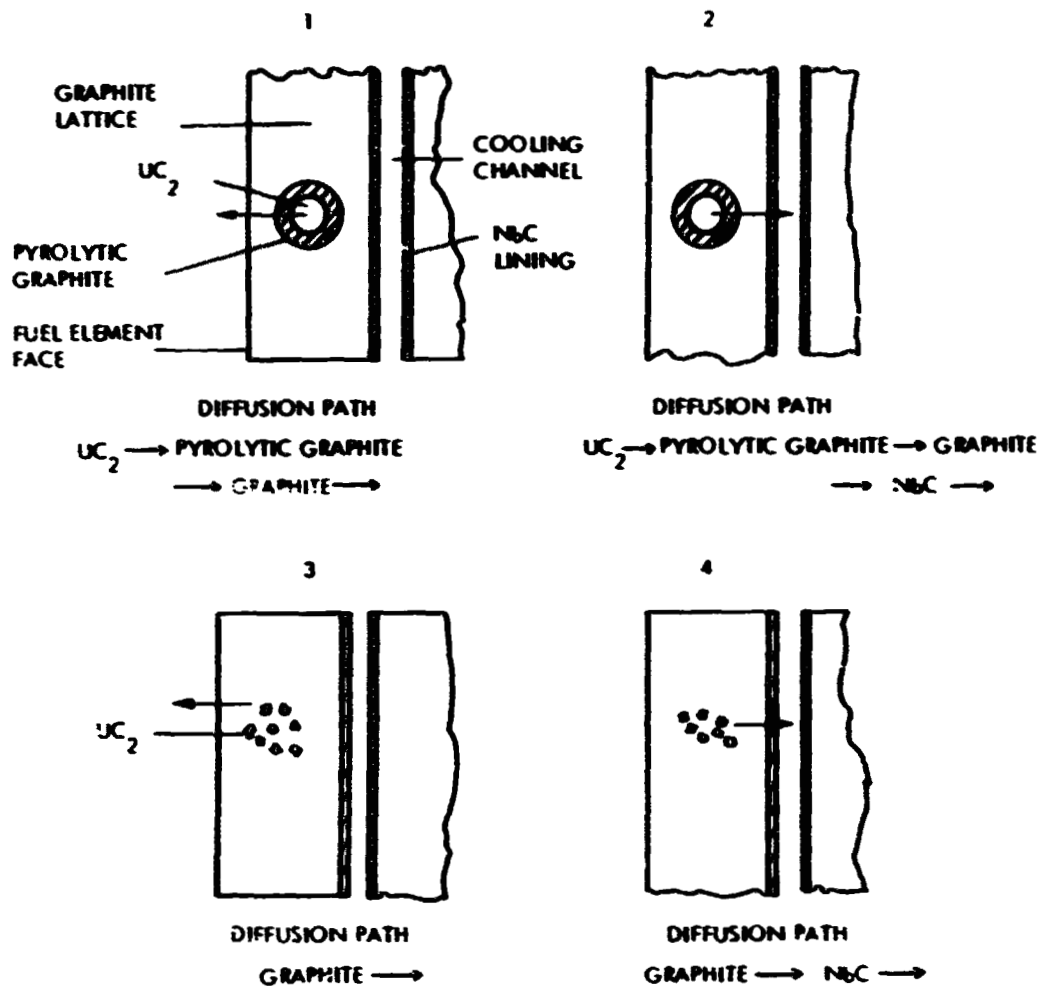
Figure 11. Fission product diffusion paths in UC$_2$ Fuel.
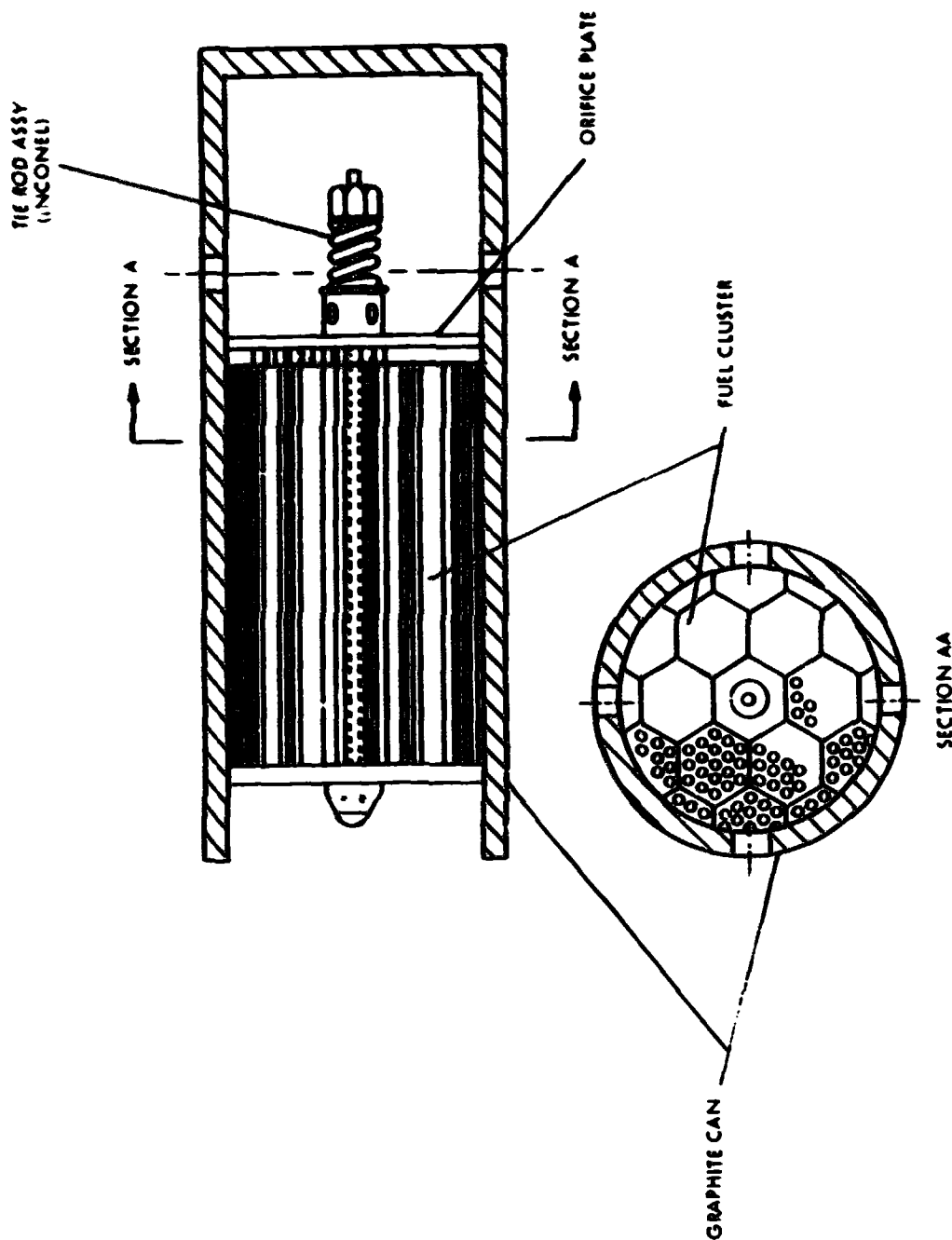
Figure 12. Cross sectional view of a Unit Cell.

TIE ROD ASSY
(INCONEL)

ORIFICE PLATE

SECTION A

SECTION A

FUEL CLUSTER

SECTION AA

GRAPHITE CAN

617753-14B
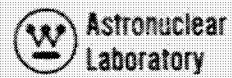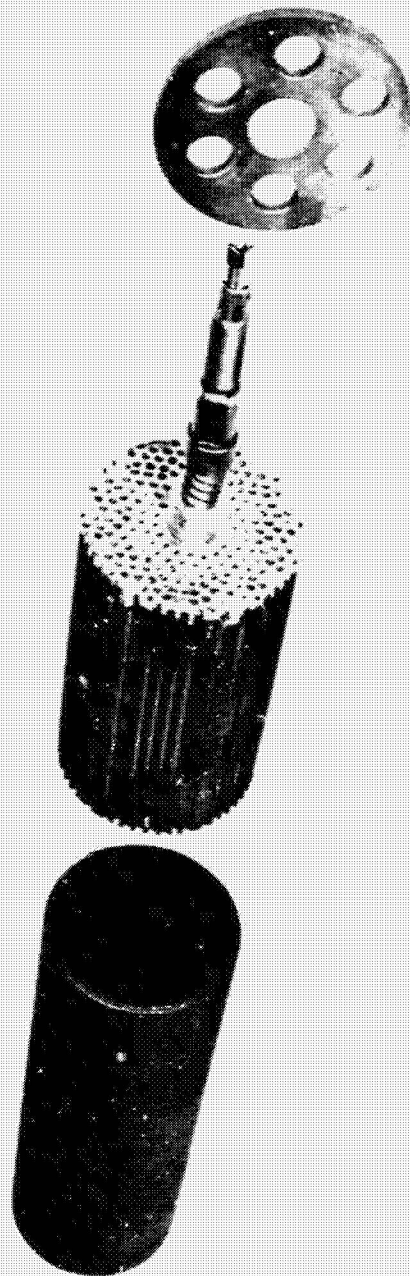
Figure 13. Exploded view of a Unit Cell assembly.

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TABLE VIII. Components of a test cell.

Item Number	Component Description	Number of Like Components	Component Dimensions and Configuration	Material
1	Fueled Element	6	Regular hexagonal, side 0.437 inch, length 4 inches - nineteen 0.097 inch diameter coolant holes.	100μ diameter UC₂ beads with 35μ diameter coating of laminar pyrolytic graphite, in graphite matrix NbC lining in coolant holes.
2	Fueled Element	6	1/2 of a regular hexagon, 4 inches long, with eleven 0.097 inch diameter coolant holes	100μ diameter UC₂ beads with 35μ diameter coating of laminar pyrolytic graphite, in graphite matrix NbC lining in coolant holes.
3	Fueled Element	6	1/4 of a regular hexagon, 4 inches long with five 0.097 inch diameter coolant holes.	100μ diameter UC₂ beads with 35μ diameter coating of laminar pyrolytic graphite, in graphite matrix NbC lining in coolant holes.
4	Unfueled Element	1	Regular hexagon, side 0.437 inch length 4 inches with 1 axial hole.	Graphite, with pyrolytic graphite sleeve.
5	Bottom Support	1	Disc, 2-3/4 inches diameter, 1/8 inches thick provided with 222 coolant holes, 0.097 inch in diameter.	Graphite
6	Top Support Plate	1	Disc, 2-3/4 inch diameter, 1/4 inch thick provided with 222 cooling holes 0.097 inch in diameter with orifice jets inserted.	Silver, Indium, Cadmium, Stainless Steel
7	Tie Rod Assembly	1	Length 9.5 inches, diameter 0.111 inch plus assorted hardware.	Inconel
8	Graphite Can	1	Cylindrical tube, closed at one end, inside diameter, 2-3/4 inch, outside diameter, 3.03 inches length 11-1/2 inches.	Graphite

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successful. Cells 2 and 3 were heated at 2300 $^{\circ}$C for 15 and 30 minutes, respectively, and Cell 4 was heated at 2450 $^{\circ}$C for 30 minutes. Each unit cell was analyzed radiochemically, following the thermal anneal, to determine release fractions of selected nuclides. The results are given in Tables XIV, XV, and XVI.

(U)   The relative magnitudes of fission product release from Unit Cells 2 and 3, both heated at 2300 $^{\circ}$C, appears incongruous since greater release was obtained from the 15-minute run than the 30-minute run. This incongruity can be resolved if the experimental heating profiles given in Figure 14 are considered. Unit Cell 2 required approximately 28 minutes to reach thermal equilibrium at 2300 $^{\circ}$C where it was held for 15 additional minutes. Since diffusion is a time-temperature dependent phenomena, fission product release will have occurred from Unit Cell 2 during the long period required to reach thermal equilibrium. Unit Cells 3 and 4 were heated differently. The furnace was brought up to temperature prior to the insertion of the unit cell assembly. With this method the sample reached thermal equilibrium in about 90 seconds. The unit cell experimental methods and results have been discussed in considerable detail in References (4) and (5).

Table XIV. Fractional release of fission products from Unit Cell 2 (15 minute thermal anneal at 2500 °C).

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 1/4 Hexagonal Fuel Elements	Average Fractional Release from Fuel	Fraction Retained by Unfueled Center Element	Fraction Retained by Outer Graphite Can	Effective Release from Unit Cell
Sr⁹⁰	0.74	0.86	0.88	0.81	0.03	0	0.77
Zr⁹⁵	0.04	0.06	0.06	0.05	0.03	0	0
I¹³¹	0.62	0.60	0.62	0.61	0	0	0.61
La¹⁴⁰	0.50	0.46	0.46	0.52	0.02	0	0.34
Ba¹⁴⁰	0.59	0.71	0.74	0.66	0.15	0.01	0.50
Ce¹⁴¹·¹⁴⁴	0.03	0.32	0.20	0.10	0.03	0.06	0.06
Gross γ Inventory	0.51	0.38	0.40	0.36	---	---	0.36

TABLE XV. Fractional release of fission products from Unit Cell 3 (30 minute thermal anneal at 2300 °C).

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 1/4 Hexagonal Fuel Elements	Average Fractional Release from Fuel	Fraction Retained by Unfueled Center Element	Fraction Retained by Outer Graphite Can	Effective Release from Unit Cell
Sr^{89}	0.89	0.94	0.94	0.92	0	0.09	0.83
Zr^{95}	0	0	0	0	0	0	0
I^{131}	0.22	0.31	0.36	0.28	0	0	0.28
Ba^{140}	0.57	0.73	0.82	0.67	0.02	0.24	0.41
$Ce^{141-144}$	0.05	0.15	0.22	0.11	0.01	0.05	0.05
Y^{91}	0.02	0.03	0.11	0.04	0.01	0.02	0.02
Gross γ Inventory	0.25	0.32	0.36	0.29	0.01	0.08	0.21

TABLE XVI. Fractional release of fission products from Unit Cell 4 (30 minute thermal anneal at 2850 °C).

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 1/4 Hexagonal Fuel Elements	Average Fractional Release from Fuel	Fraction Retained by Unfueled Center Element	Fraction Retained by Outer Graphite Can	Effective Release from Unit Cell
Sr	0.91	0.97	0.98	0.96	0.01	0	0.95
Zr	0.03	0.02	0.07	0.03	0	0	0.03
I131	0.30	0.35	0.33	0.32	0	0	0.32
Ba140	0.48	0.77	0.83	0.65	0.02	0.12	0.51
Ce141	0	0.11	0.21	0.08	0.03	0.06	0.01
Y91	0.06	0.11	0.18	0.10	0	0.01	0.09
Mo99	0.02	0.04	0.06	0.03	0	0	0.03
Te132	0.23	0.24	0.57	0.26	0	0.01	0.25
Gross γ Inventory	0.21	0.30	0.34	0.26	0.01	0.07	0.39

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Figure 14. Heating profiles for Unit Cells 2 and 3.

B.   .omparison between Calculations and Experimental Results

(U)   Diffusion constants, derived from laboratory experiments with small
sections of degraded fuel, were used in the FIPDIF computer program to predict
release fractions from NERVA fuel elements. For the simulated loss-of-coolant
profile experiment, the predicted fractional release of 0.673 for gross gamma
activity agreed very well with the experimental results of 0.667. Uncoated
Type III diffusion constants were used for the prediction and a single section
of fuel 1-1/2 inches long was used for the experiment. Although the agreement
was very good in this experiment, the question remained whether these data would
be applicable to the escape of fission products from the reactor proper.
(U)   The results obtained from the unit cell experiments showed that the
above method could not be used to predict release from the reactor proper because
the measured release from the unit cells was appreciably smaller than the FIPDIF
predictions using Type III diffusion constants. A better comparison between
predictions and results was obtained when Type III-NbC diffusion constants were
used in the FIPDIF program. A sample of this comparison is given in Table XVII
for Sr^{89} and Ba^{140}. The predicted release values were significantly lower and
agreed much better with the radiochemical analysis when Type III-NbC constants
were used. This finding tends to reinforce the hypothesis that the release of
fission products from the NERVA reactor proceeds by diffusion through the NbC
coating in the fuel element cooling channels. In general, however, the release
values determined radiochemically were still lower than the predictions using
Type III-NbC diffusion constants. Other factors such as coolant channel blockage
or exchange of fission products between adjacent fuel elements may be responsible
for these results.

TABLE XVII. Comparison of measured fission product release ... peak inlet ... with predicted release.

Cell Number and Experimental Conditions	% Xe Release Radiochemical Determination	% Xe Release Predicted with Type III Diffusion Constants	% Xe Release Predicted with Type III-NRX Diffusion Constants	% Release Radiochemical Determination	% Release Predicted with Type III Diffusion Constants	% Release Predicted with Type III-NRX Diffusion Constants
2 Heated 2300°C for 15 minutes, 20 minutes to achieve temp. equilibrium	77	100	67	50	100	47
3 Heated to 2300°C for 30 minutes, 1.5 minutes to achieve temp. equilibrium	83	100	85	41	100	72
4 Heated to 2450°C for 30 minutes, 1.5 minutes to achieve temp. equilibrium	95	100	97	50	100	83

C. Latest Diffusion Constants

(U)   During a loss-of-coolant condition, the core temperature rises rapidly above the degradation point of the fuel. To predict the amount of fission products released, the FIPDIF code requires diffusion constants of fission products from degraded or Type III fuel. Such constants were derived from release studies at 2300, 2400, and 2500 $^{\circ}$C for Y, Zr, Ru, Sb, Te, I, La, and Ce. Similar constants for Sr, Ag, Sn, Xe, Cs, and Ba have been determined earlier on Type III NERVA fuel[4] and were considered applicable.

(U)   The diffusion constants for Rb were determined by assuming release rates at two temperatures corresponding to that of a medium-fast diffuser and using these values to compute a frequency factor and an activation energy. The release measurement of Rb is extremely difficult because of the fact that the only nuclide with a reasonably long half life, 18.6-day Rb[86], is a shielded nuclide of low fission yield.

(U)   The diffusion constants intended for FIPDIF predictions above the degradation point of A6 type fuel are presented in Table XVIII.

TABLE XVIII. Summary of diffusion constants for
selected nuclides in degraded A6 fuel.

Nuclide	D_o (Sec^{-1})	E/R(^{o}K x 10^3)
Rb	400	15
Sr[1]	1.63	11.4
Y	4.01 x 10^7	70.6
Zr	0	0
Mo	0	0
Ru	0	0
Cd	400	15
Ag[1]	1	20
Sn[1]	18.4	18.2
Sb	.78	17
Te	649	38.7
I	7.47 x 10^8	76.7
Xe[1]	1.63	11.4
Cs[1]	0.48	10.5
Ba[1]	1800	26.2
La	21.6	25.0
Ce	0.78	17

[1]These constants are derived from release measurements on
Type III NERVA fuel, see Reference 4.

IV. RECENT EXPERIMENTS

A. Ag[111] Release Measurements

(U)   Most fission products do not diffuse significantly from Triplex Fuel at temperatures below 1800 °C. An exception to this is silver which diffuses more rapidly than any of the fission products studied. Since nearly all of the silver was released in the higher temperatures experiments, separate experiments were performed at lower temperatures to determine the diffusion characteristics of this nuclide. The diffusion of this species is important because it is a precursor to cadmium which is used to determine aliquot factors of effluent samples from reactor testing. Although diffusion experiments for silver were run on all types of fuel, only the most recent experiment involving NbC-coated Triplex Fuel is described here.

(U)   Irradiated samples of Triplex Fuel, 1/4-inch thick, completely coated with NbC, were heated at temperatures of 1200 °C, 1400 °C, and 1600 °C for time periods of 5, 15, 30 and 60 minutes. The release of Ag[111] from each sample was determined by radiochemical analysis. Diffusion constants were calculated for each of the three temperatures and were plotted against $1/T$ °K to determine the overall diffusion constant. The results of this experiment are shown in Figure 15 along with an error envelope calculated at the 95 percent confidence limit. Similar curves for other nuclides have been reported previously.[6]
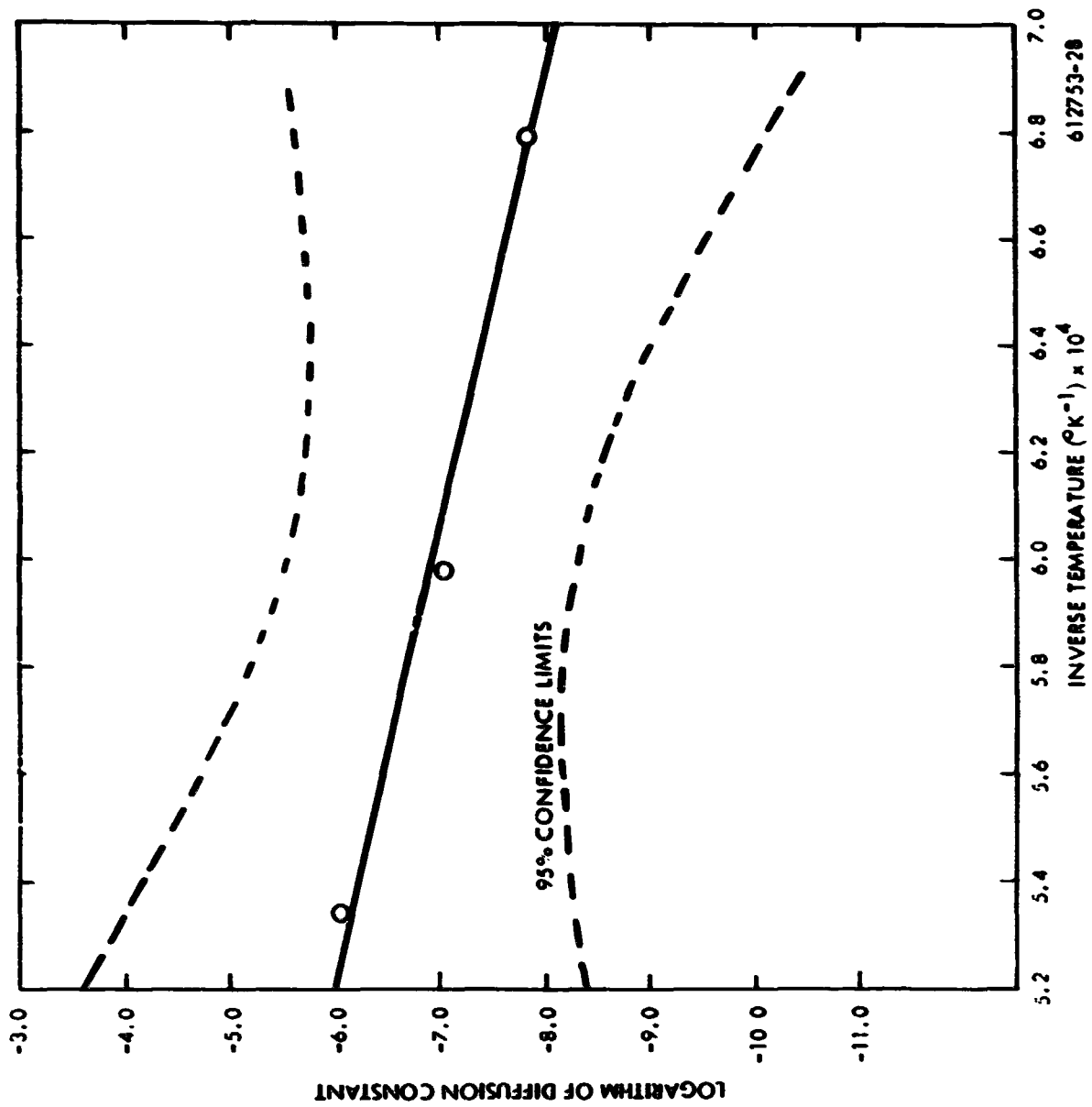
Figure 15. Arrhenius plot of the release of Ag^{111} by diffusion from NbC Triplex Fuel.

B. Release of Te^{129m} during NRX-A6 Tests

(U)   The release of fission product activity during NRX-A6 testing was determined by analysis of effluent samples taken during testing and by post test fuel analysis. The effluent samples were taken at 5, 12, and 20 minutes from the beginning of full power operation. At this point in time the sampling apparatus, the 'Elephant Gun', malfunctioned, precluding further sampling. The fuel analyzed consisted of fifty 1/4-inch slices, representing a five by ten matrix of the NRX-A6 core. Ten equally spaced slices were taken from each of five fuel elements selected from different radial positions in the core. The results of these analyses were reported previously[11] except for 37-day Te^{129m} which was completed only after the report had been issued. The measurement of Te release was undertaken to derive diffusion constants of Te isotopes which are the precursors of the biologically important iodine isotopes.

(U)   The results of the fuel analysis for Te^{129m} showed that 9.42 percent of mass chain 129 was released during the 60-minute operation of NRX-A6. With this relatively high release of Te^{129m}, the 'Elephant Gun' data was examined for Te^{129m} content. The tellurium activity found in the 'E' gun samples was originally assumed to be all 78-hour Te^{132}. An examination revealed that 25 percent of the tellurium activity in the 'E' gun counting samples was Te^{129m}. The counting data was resolved for the two components and calculations were made to determine the release values of both Te^{129m} and Te^{132}. These results are given in Table XIX along with fuel analysis results, and the most recent FIPDIF computer program release predictions for these nuclides. The results show good agreement between fuel analysis and the FIPDIF predictions and fair agreement with the effluent results. This is also consistent with laboratory data. Experiments in the laboratory have shown that tin diffuses faster than antimony, and antimony diffuses faster than tellurium through NERVA fuel. These elements are members of mass chain 129 and 132 which are shown in Figure 16 with their respective half lives and fission yields.[13] By analyzing these chains it can

TABLE XIX. Tellurium activities released in NRX-A6 tests.

ELEPHANT GUN

Sample	Original Te^{132} Release (%)	Corrected Te^{132} Release (%)	Te^{129m} Release (%)
5 Min	0.735	0.546	4.68
12 Min	0.971	0.720	6.19
20 Min	1.08	0.804	6.88
Composite	0.950	0.706	6.05

FUEL ANALYSIS - 60 MIN. RUN TIME

Te^{129m} Release = 9.42%

Te^{132} Release --- Determination not possible because of natural decay.

FIPDIF PREDICTIONS - 60 MIN. RUN TIME

Te^{129m} Release = 9.87%

Te^{132} Release = 0.45%

be seen that during a 60-minute run most of the mass 129 atoms are present as
Sn^{129} and Sb^{129} atoms, while most of the atoms in the mass 132 chain are present
as Te^{132} atoms. Since tin is the fastest diffuser in these chains, it follows
that much more release should occur in the mass 129 chain than in the mass 132
chain.
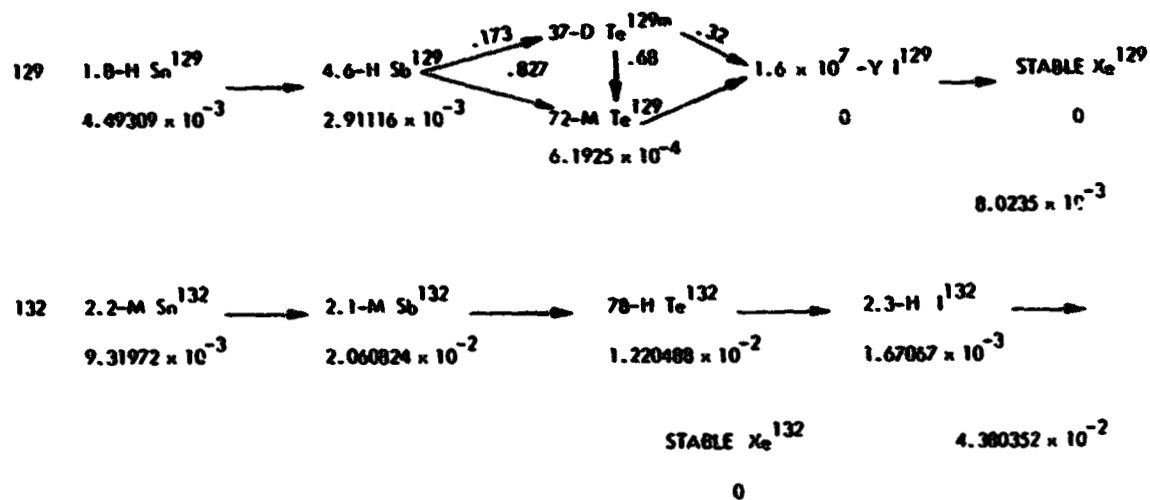
FISSION YIELDS OF MASS CHAINS 129 AND 132   612753-168

Figure 16. Fission yields of mass chains 129 and 132.

C. Experimental Approaches to Prepare Degraded Fuel

(U)   Two experimental methods were considered for the determination of diffusion constants for the loss-of-coolant conditions. One method was to irradiate intact fuel and heat the samples at temperatures above the degradation point. The other method was to degrade the fuel before irradiation and then heat at lower temperatures. A preliminary experiment was performed to determine which of the two methods would be used for further experimentation.

(U)   Sixteen, 1/4-inch samples were used from a regular, channel coated, Triplex Fuel element for this experiment. Eight of these samples were kept intact, while the other eight were preconditioned by heating in an argon atmosphere at 2500 $^{\circ}$C for 15 minutes. After irradiation, twelve of the samples were heated, individually, for 15-minute time periods at temperatures of 2300 $^{\circ}$C, 2400 $^{\circ}$C and 2480 $^{\circ}$C. The latter temperature was originally scheduled for 2500 $^{\circ}$C, but 2480 $^{\circ}$C was the maximum temperature that could be achieved without changing the tap positions in the RF generator. The other four samples, two of each type, were retained as controls. All of the post-irradiation thermal anneals were done in a flowing helium atmosphere to minimize graphite sublimation. The gross gamma and La^{140} release values were obtained by gamma counting the samples before and after each heat. The samples were then dissolved and analyzed radiochemically for Y^{91}, Zr^{95}, Mo^{99}, I^{131}, Te^{132}, and Ce^{141}. Results from the 2300 $^{\circ}$C temperature were scattered because release values were low. However, good data was obtained from the 2400 $^{\circ}$C and the 2480 $^{\circ}$C runs. The results, given in Figures 17 and 18, clearly show that fission product release is much less for intact fuel than it is for preconditioned fuel. Because the fuel is initially intact for the loss-of-coolant situation, the decision was made to determine degraded diffusion constants by using intact fuel and heating at temperatures above the degradation point, even though it is the more difficult of the two methods.
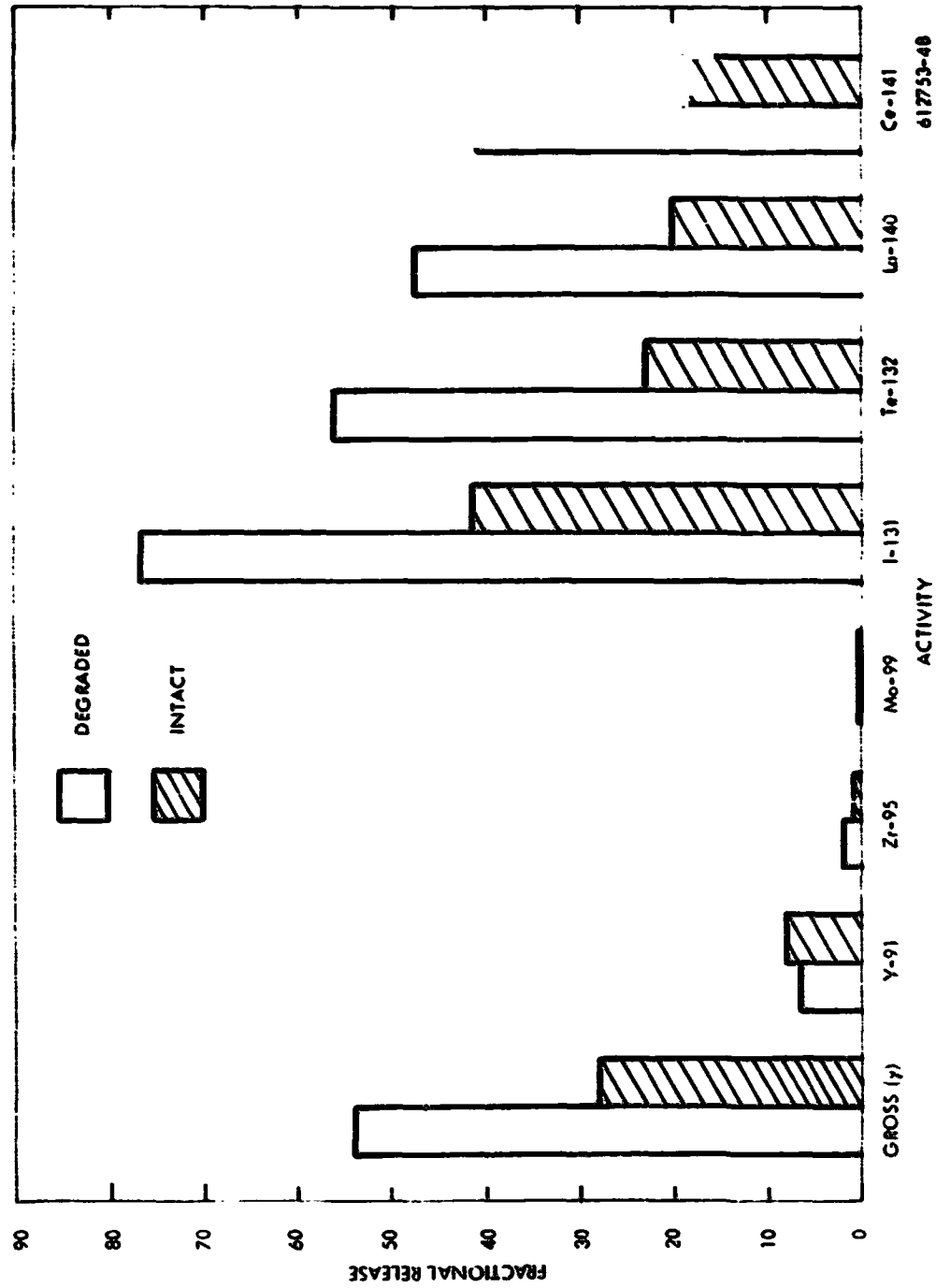
Figure 17. Fission product release from Triplex Fuel after heating
for 15 minutes at 2400 °C in helium.
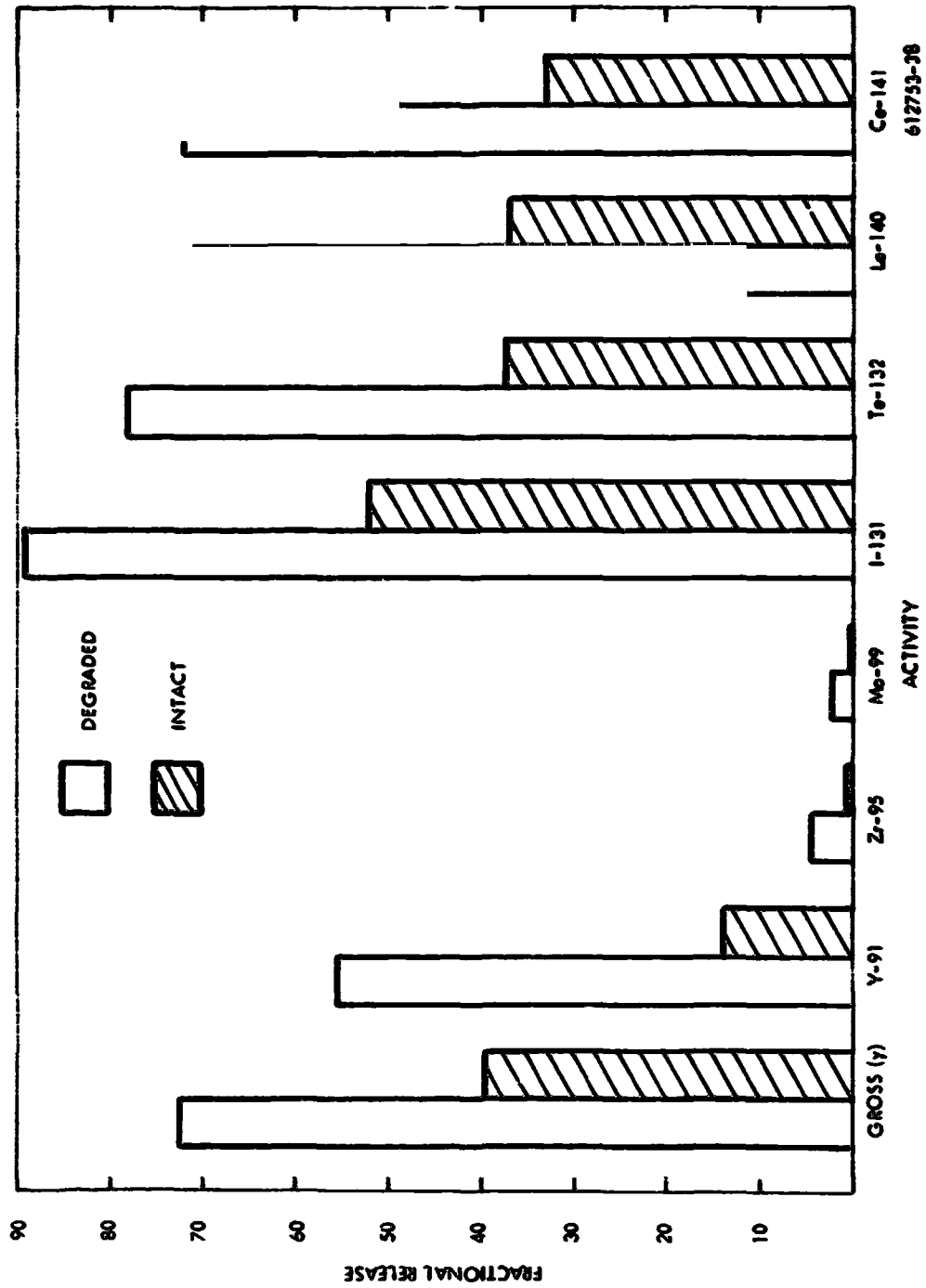
Figure 18. Fission product release from Triplex Fuel after heating for 15 minutes at 2480 °C in helium.

D. Fission Product Release from Triplex Fuel Beyond the Degradation Point

(U)   Three separate experiments were performed to determine fission product
release for the loss-of-coolant situation. Fourteen, 1/4-inch slices of produc-
tion type Triplex Fuel were used for each experiment. Twelve of these samples
were subjected to post-irradiation thermal anneals and two were retained as
control samples. The thermal anneals consisted of heating the samples, individually,
at temperatures of 2300 ^{0}C, 2400 ^{0}C, and 2500 ^{0}C for time periods of 5, 10, 20
and 30 minutes. Each sample was heated in a flowing helium atmosphere. The gross
gamma fission product release was determined by gamma counting the samples before
and after each heat. The release of La^{140} was determined by counting the samples,
before and after each heat, on a 512 channel gamma ray spectrometer and observing
the change in the 1.60 MeV gamma peak. The fuel slices, including the controls,
were then dissolved and analyzed radiochemically for Y^{91}, Zr^{95}, Mo^{99}, Ru^{103}, Sb^{127},
Te^{129m}, I^{131}, and Ce^{141}. The fractional release of each nuclide was determined by
comparing the activity in the heated sample with that in the controls. The results
of the three experiments were averaged and are given in Table XX. Smooth data was
obtained in most cases. The exceptions were Zr^{95}, Mo^{99}, and Ru^{103} where low release
values caused a fluctuation in the final results. The release was less than 10
percent in all cases for these nuclides.

TABLE XX. Release fractions of fission products from Triplex Fuel.

Temp. Deg. C	Time (Min.)	Y^{91}	Zr^{95}	Mo^{99}	Ru^{103}	Sb^{127}	Te^{129m}	I^{131}	La^{140}	Ce^{141}	Gross Gamma
2300	5	.048	.039	.008	.001	.086	.001	.126	.062	.070	.075
	10	.085	.004	.032	.001	.124	.114	.194	.100	.101	.130
	20	.105	.029	.011	.034	.200	.108	.212	.147	.153	.161
	30	.114	.041	.006	.019	.158	.151	.202	.200	.199	.174
2400	5	.060	.038	.001	.065	.135	.068	.356	.120	.130	.130
	10	.103	.031	.028	.025	.362	.265	.552	.287	.256	.294
	20	.210	.022	.026	.069	.494	.446	.615	.478	.438	.424
	30	.283	.037	.026	.031	.616	.538	.687	.646	.594	.522
2500	5	.087	.016	.034	.032	.346	.282	.583	.285	.236	.295
	10	.179	.025	.011	.055	.541	.507	.726	.514	.441	.445
	20	.323	.063	.068	.073	.704	.673	.806	.721	.647	.582
	30	.428	.042	.088	.005	.766	.740	.866	.836	.770	.654

V. SOURCE TERM PREDICTIONS FOR R-1

A. Normal Operation

(U)   The FIPDIF prediction of the release in percent of selected fiss'on
products during a one-hour run of the R-1 reactor at full power is summarized in
Table XXI. The power profile was assumed to be of a simple square wave shape.
The temperature profile of the R-1 core at full power was taken from Reference 15.
For comparison, the FIPDIF prediction for the A6 reactor run is also included in
Table XXI. The power output of A6 increased during the first ten minutes from
zero to full power and was kept at this level for 60 minutes. The diffusion
constants used for the FIPDIF predictions were the same in both cases, i.e., the
R-1 fuel was taken to be the same as the A6 fuel.

(U)   Various safety organizations are particularly concerned about the release
of iodine activities due to its biological significance. A comparison between
the release of mass chains 131 to 135 from R-1 and A6 is presented in Table XXII.

(U)   Since the R-1 is designed for operation at higher core temperatures than
the A6, it is not surprising that FIPDIF predicts a higher release of fission
products both in terms of number of atoms and percent. The conceptual difficulties
as to the exact meaning of 'gross gamma' are discussed in Chapter VII.

TABLE XXII. Comparison between the computed percent release of selected mass chains from R-1 and A6 reactors after 60 minutes of full power operation.

Mass Chain	Percent Release from	
	R-1	A6
89	4.27	2.97
91	1.40	0.89
95	0.056	0.036
111	33.0	33.7
115	51.6	47.7
123	20.7	20.6
131	2.14	1.26
137	5×10^{-3}	8×10^{-4}
140	1.30	0.94
141	2.29	1.56
144	2.01	1.30
Gross Gamma	1.26	0.84

TABLE XXIII. Predicted iodine and gross gamma release from A6 and R-1.

Nuclide	A6 Release		R-1 Release	
	Percent	Curies	Percent	Curies
I-131	1.26	1.44×10^3	2.14	3.11×10^3
I-132	0.44	6.25×10^4	0.67	1.21×10^5
I-133	0.07	1.61×10^3	0.38	1.15×10^4
I-134	0.05	2.65×10^4	0.26	1.85×10^5
I-135	0.08	5.75×10^2	0.45	4.16×10^4
Gross Gamma	0.84	2.30×10^7	1.26	2.84×10^7

B. Loss of Coolant Accident

(U)   For the prediction of the fission product release during a loss-of-coolant accident, the R-1 was assumed to be fueled with A6 type fuel and to operate normally for 30 minutes at its design power level of 1575 megawatts after which a loss of coolant was assumed to occur lasting for 10 minutes before loss of structural integrity. The power profile during normal operation again was assumed to be of a simple square wave shape. The core temperatures required as input data for FIPDIF were extrapolated from a theoretical analysis of post-operative heating of the R-1.[16] The predicted release of some fission products for normal and accident cases are presented in Table XXIII.

TABLE XXIII.   Predicted release of selected fission products from R-1 after 30 minutes of operation followed by a loss-of-coolant failure.

	Percent Release After	
Mass Chain	30 Min. Normal Operation	Loss of Coolant
89	2.52	22.3
91	0.92	7.9
95	0.048	0.1
111	17.6	38.7
115	46.7	54.2
123	12.3	44.7
131	1.75	69.9
137	0.005	0.6
140	0.65	6.7
141	1.01	17.3
144	1.07	20.2
Gross Gamma	0.67	18.2

(U)   Two facts are immediately obvious from an inspection of Table XXIII:

1. Nuclides which under normal operating conditions would not measurably diffuse such as Zr^{95} or Cs^{137}, are being released under accident conditions, and

2. The fraction of I-131 which is expected to be released is substantial.

VI. EFFLUENT SAMPLING TECHNIQUES

(U)   The release by diffusion of fission product activity was determined by
post-operational fuel analysis as described in Chapter II. Although fuel analysis
is important and yields valuable information, it does leave something to be
desired. For instance, fuel analysis cannot resolve total release values into
release values for separate full-power runs of a single reactor. Also, it cannot
determine the fission product release due to corrosion. Another disadvantage of
fuel analysis is that for low release nuclides small errors in radiochemical
analyses produce large errors in the final release values. To obtain more
extensive release data, and in some cases, more reliable diffusion data, methods
for sampling the reactor effluent during full-power operation were employed.

(U)   One method was the use of aircraft, with wing-mounted filters, to sample
the exhaust gas cloud during, and immediately after full-power operation of a
reactor. This method was employed for the full-power runs EP-4 and EP-4A of the
NRX-A4 reactor, and for EP-3 and EP-4 of the NRX-A5 reactor. In each case the
filters were retrieved, dissolved, and analyzed radiochemically. Cloud aliquot
factors were determined by using Cd^{115} as an indicator nuclide. This nuclide
was chosen because its release value during full-power operat.   is near 50
percent, and its exact value can be determined quite accurate: fuel analysis.
The fission product loss from the reactor due to corrosion was determined by
analyzing the aircraft filter samples for Mo^{99}. This nuclide was chosen because
laboratory data shows that none of the elements in mass chain 99 diffuse under
normal reactor test conditions. Therefore, any Mo^{99} activity observed in the
filter samples was presumed to be from corrosion and not diffusion. With this
analytical tool it was possible to resolve total release values into the two
components, release by diffusion and release by corrosion.

(U)   To illustrate the usefulness of aircraft sampling, the release values
of some selected nuclides for separate full-power runs on NRX-A4 and NRX-A5 are
given in Table XXIV. These values are given in terms of curies at time of

TABLE XXIV Fission product activity from NRX-A4 and NRX-A5 based on aircraft filter samples.

Nuclide	Curies Released From NRX-A4			Curies Released From NRX-A5		
	EP-4	EP-4A	Total	EP-3	EP-4	Total
Sr-89	335	2420	2755	71	920	991
Zr-95	322	1580	1902	3	1100	1103
Ag-111	80	113	193	44	70	114
I-131	936	4020	4956	170	1100	1270
Ba-140	866	6270	7136	80	2800	2880
Gross Gamma	1.2×10^7	7.0×10^7	8.2×10^7	0.3×10^7	3.7×10^7	4.0×10^7

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shutdown and include release by diffusion and corrosion. Calculations were also
made for these tests to resolve combined release values into their diffusion and
corrosion components. An example of this is given in Table XXV for gross gamma
release in terms of percent of total inventory. More detailed results of release
from NRX-A4 and NRX-A5 have been reported in References (9) and (10). Aircraft
filter analysis provided a great deal more data with much less effort than did
fuel analysis. Discrepancies were observed, however, when release values by
diffusion were compared between aircraft filter and fuel analyses. In many cases
much higher values for release by diffusion were obtained from filter analysis
than from fuel analysis. A possible reason for this could be the retention
efficiency of the filter material for different nuclides in the effluent cloud.
Therefore, to obtain better diffusion data, a new device for sampling the effluent,
designated 'Elephant Gun', was developed by the Los Alamos Scientific Laboratory.

(U)   The 'Elephant Gun' consists of a revolving-type drum with eight cylinders,
a stainless steel canister positioned in each of the eight cylinders, and a
delivery tube through the test cell concrete shield wall. The system is designed
to sample the effluent gas stream just above the reactor exhaust nozzle. At
selected time periods the canisters are injected into and extracted from the
effluent stream, one at a time. This permits eight samples of effluent gas to
be taken during the course of the run. The 'Elephant Gun' was made available to
WANL for sampling effluent during the NRX-A6 test. Unfortunately, only three
samples out of the planned eight were obtained because of a system malfunction.
This provided fission product release data only up to 20 minutes of full-power
operation. The reactor was operated at full power for a total of 60 minutes.
Consequently, a direct comparison of release values could not be made between
'Elephant Gun' samples and fuel analysis. The results were reasonable, however,
since the canister analysis from 20 minutes of full-power operation gave lower
release values than fuel analysis. The fuel samples experienced the full 60-
minute run time. The 'Elephant Gun' sampling system was concluded to be a worth-
while supplement to the fuel analysis. The complete results of fission product
release from NRX-A6 have been reported in Reference (11).

TABLE XXV. Gross gamma release from NRX-A4 and NRX-A5
based on aircraft filter samples.

Reactor	Experimental Plan	Percent Release of Total Inventory		
		Diffusion	Corrosion	Total
NRX-A4	EP-4	0.80	0.65	1.45
	EP-4A	2.95	4.41	7.36
NRX-A5	EP-3	0.29	0.01	0.30
	EP-4	1.70	2.30	4.00

VII. ACCURACY OF SOURCE TERM PREDICTION

A. Predicted Source Terms

(U)   The designation 'source term' has had several meanings in conjunction with the NERVA Program. In its broadest sense source term refers to the amount of fission product generation and decay in reactor fuel, the diffusion of fission products from the fuel at high temperatures, neutron activation of NERVA engine materials and the effects of re-entry oxidation and ablation of reactor fuel and structural materials on the total radioactive inventory.

(U)   The source term which is used in conjunction with the fission product diffusion program is more restricted.

(U)   For our program, source term is defined as the quantity of any radio-active fission product remaining in the NERVA reactor plus the quantity of that nuclide released from the reactor during reactor operation.

(U)   As an example, the I^{131} source term for a particular reactor run is the total number of curies of I^{131} remaining in the core at T_1, which is the reactor shutdown time, together with the total number of curies of I^{131} released up to time T_1. In practice, the activity released is that of mass chain 131, and for short periods after the run the activity would be due to iodine precursors Sn^{131}, Sb^{131}, and Te^{131}. Thus the I^{131} source term may be considered as potential I^{131} calculated at time of reactor shutdown.

(U)   A special case which must be considered is the gross gamma source term. In this instance we are concerned with the sum of the activities of all nuclides which emit gamma radiation. The relative proportions of the various nuclides contributing to the gross gamma activity changes with time. For time periods of the order of minutes after a fission event the change in the relative propor-tions of the contributing nuclides is very rapid. All of the gross gamma source term predictions to date have been calculated at time of reactor shutdown. It

must be appreciated that these calculated gross gamma source term predictions
will not exactly correspond to the measured gross gamma activity at any time
after reactor shutdown.

(U)   For all of these predicted source terms the calculations are made with
a computer code called FIPDIF. FIPDIF is a program written in Fortran IV for
the calculation of the fission product release and the fission product inventory
remaining in a multi section NERVA reactor core. In its present form it can be
used to simulate a reactor startup-run-shutdown profile consisting of as many as
forty time intervals. These time intervals may be of any desired length. During
each time interval, a power level and temperature is specified for each core
section. As many as sixteen core sections may be used. At the conclusion of any
or all of the specified time intervals, the following data may be printed out:

(U)   a. The remaining fission product inventory in each core section and/or
the total core inventory of each isotope.

(U)   b. The curies of each or all isotopes which have diffused from each
section and/or the total curies of each or all isotopes which have diffused from
the core.

(U)   A more complete descritpion of this code is given in Reference 17.

B. Measured Source Terms

(U)   The quantities of radioactive nuclides released from the reactor or
retained in the core have been determined by several techniques described in
Chapters II, III and IV of this report. The quantity of any nuclide measured
subsequent to a reactor test can be corrected for decay to reactor shutdown time
by well established decay constants. Thus, a normalization procedure can be made
for making correct comparison of measured release with predicted release.

(U)   A special case is the gross gamma source term. Measurements of the
gross gamma activity released from the reactor or retained in fuel slices are
made in an ionization chamber. Extrapolation of these measurements back to
reactor shutdown time is made with the Way-Wigner relationship (Chapter II) which
gives the activity of mixed fission products as a function of time. This
relationship is calculated, rather than experimental, with an estimated accuracy
of 20 percent. Thus, the measured gross gamma source term is less accurate than
the measured source term of individual nuclides.

C. Comparison of Predicted and Measured Source Terms

(U)   In order to test the accuracy of the source term predictions for the release of fission products, ratios of the predicted source terms to the measured source terms for the various reactor tests have been calculated. In this manner the ratio becomes an accuracy index, with accuracy increasing as the value of the ratio approaches unity. Conservative source terms, with the predicted release of fission products greater than the measured release, will result in ratios greater than unity.

(U)   The ratios of the predicted I^{131} source term to the measured I^{131} source terms for the various reactor tests are summarized in Table XXVI. It may be seen that a minimum ratio was 0.44 for the NRX-A3 test, while the maximum ratio was 1.97 for NRX-A6.

(U)   The discrepancy between predicted and measured release in the case of NRX-A3 is believed due to too low a value of the predicted release. For this calculation appropriate diffusion constants were not used. As a result of this discrepancy the diffusion constants were modified and the predicted and measured I^{131} source terms of the next reactor test, NRX-EST EP-IV A, were nearly identical.

(U)   The discrepancy between measured and predicted I^{131} source terms for the NRX-A6 test is believed due to an incorrect measurement of the I^{131} release. Values of release for the NRX-A6 reactor were determined by an insensitive technique of post mortem fuel analysis. The small value of I^{131} release from NRX-A6 is inconsistent with all previous runs and therefore suspect.

(U)   The minimum and maximum ratios of predicted and measured source terms indicate that errors exist in the measurements as well as the predictions. In view of combinational probabilities of these errors, discrepancies in the 'Predicted over Measured' ratios for individual nuclides of the order of 100 percent can be expected, especially when the release is low.

(U)   Included in Table XXVI are ratios of predicted-to-measured gross gamma source terms. The use of improved diffusion constants have also improved these predictions to the point where prediction and measurement compare favorably.

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TABLE XXVI. Comparison of predicted and measured Source Terms.

Reactor Test	Predicted* I131 Release (% of Total I131 Inventory)	Measured I131 Release (% of Total I131 Inventory)	Predicted I131 / Measured I131	Predicted* Gross Gamma Release (% of Total Gross Gamma Activity)	Measured Gross Gamma Release (% of Total Gross Gamma Activity)	Predicted Gross Gamma / Measured Gross Gamma
NRX-A2	1.15	1.66	0.70	1.24	1.02	1.22
NRX-A3	0.80	1.80	0.44	1.6	1.0	1.60
NRX-EST EP-IV	2.20	2.06	1.07	2.10	0.80	2.62
NRX-EST EP-IV A	2.77	5.05	0.55	2.32	2.95	0.79
NRX-A5 EP-III	0.75	0.70	1.07	0.42	0.30	1.40
NRX-A5 EP-IV	1.16	0.80	1.40	2.38	1.7	1.40
NRX-A6	1.26	0.64	1.97	.84	0.83	1.01

*Based on post-run power and temperature profiles and revised diffusion constants.

VIII. ADDITIONAL INFORMATION REALIZED FROM DIFFUSION EXPERIMENTS

A. Bead Damage Studies

(U)   The isothermal release by diffusion of most fission products from
irradiated NERVA fuel sections was found to follow a two-term rate equation of
the form

$$f = g_1 \exp(-D_1 t) + g_2 \exp(-D_2 t), \tag{3}$$

where f is the fraction of fission products retained, t the heating time, D_1,
D_2 are diffusion constants, and g_1, g_2 are weighting factors defined as $\Sigma g_i = 1.00$.
The two terms in equation (3), describing diffusion through two different barriers,
differ vastly in magnitude. The combined effects of the pyrocarbon coating around
the beads and the NbC coating of the coolant channels is believed to be accounted
for by the term $g_2 \exp(-D_2 t)$, the slowly diffusing fraction. The 'fast' term,
$g_1 \exp(-D_1 t)$, is believed to describe diffusion through the graphite matrix.
(C-RD)   Recent measurements of the Xe^{133} release from fuel beads, crushed in
various degrees suggested that the 'fast' term in equation (3) indeed is related
to the magnitude of exposed, i.e., hydrolyzable, UC_2 surfaces. Thus, the 'fast'
term describes diffusion of Xe^{133} through UC_2 and evaporation from an open UC_2
surface. In fact, the amount of Xe^{133} released, in terms of counts per minute,
under standardized experimental and counting conditions has been directly related
to the amount, in milligrams, of hydrolyzable UC_2 in a given fuel section.
Because of the wide applicability of this method, the standard experiment condi-
tions are summarized below:

a. A 1/4-inch section of fuel is irradiated to ~3 kW-hours which
corresponds to approximately 2×10^{13} fissions.

b. The exact number of fissions which have occurred in the sample is
measured with an ionization chamber.
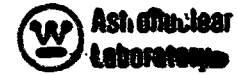
c. The sample is heated to 2000 °C for 30 minutes with a 45 to 50 second heat-up time, the time required to reach 2000 °C.

d. The released Xe^{133} is adsorbed on 2-3 grams of activated 6-15 mesh charcoal at liquid nitrogen temperature in special U-shaped tubes.

e. The heating must take place as closely to 4.0 days after the end of the irradiation as possible.

f. Each Xe^{133} sample is counted at least three times on a 3" x 3" NaI (Tl) gamma detector in a special holder 10 cm from the crystal face. The net count rate is that of the 81 keV gamma ray of 5.3-day Xe^{133} with natural background and Compton contribution subtracted.

g. The counting data is normalized to 3.275 x 10^{13} fissions per sample and to 140.0 hours after the end of the irradiation.

(C-RD) As expected, the curve relating Xe^{133} count rate with 'hydrolyzable' uranium is not a straight line. The Xe^{133} measured has two sources of origin: (a) release from broken or damaged beads, and (b) release by diffusion from intact beads. The ratio of the amounts of Xe^{133} from each source is not constant for different sample sizes. Mathematical treatment of the input data to straighten the calibration curve results in greater uncertainties. In its present form, the above method has a lower limit of detection of about one milligram of uranium, sufficiently low for the detection of exposed uranium in a production fuel element.

(This page is unclassified.)

B. Temperature Determinations Based on Diffusion Measurements

(U)   It has long been recognized that the release of fission products from NERVA fuel is a temperature-sensitive process. Also, the temperature dependence of the removal rate of fission products can adequately be described by an Arrhenius type relation which suggests that the removal mechanism is mostly diffusion. Thus, knowing the two material-dependent constants in the Arrhenius equation, frequency factor and activation energy, one should be able to deduce the temperature to which a section of fuel was exposed when the release of a suitable fission product is meas.

1. Relation Between Fractional Release and Temperature

(U)   In order to derive an analytic expression relating a measured quantity, the fractional release of a fission product with the quantity of interest, the fuel temperature at 'peak' power operations, the following conditions should be satisfied:

a. The indicator nuclide should be a slow or medium fast diffuser.

b. Axial temperature and power profiles should be of the same shape everywhere in the core.

c. Release rates of the indicator nuclide varies from location to location within the core only because the core temperatures vary; i.e., release by corrosion is excluded.

d. The temperature dependence of the release rate of the indicator nuclide is given by an Arrhenius-type equation of the form $W = R_0 (\exp{-E/T})$, where R_0 is a frequency factor and E an activation energy both of which have been determined in laboratory experiments.

e. The ratio of the fractional releases (f) at the temperatures T_1 and T_2 should be equal to the ratio of the amounts (``) released at the same temperatures, or

$$f(T_1)/f(T_2) = N(T_1)/N(T_2). \tag{4}$$

(U)   The va`` ``lity of equation (4) has been verified in laboratory experiments.

(U)   To derive the relation between fractional releases and temperatures, $R_o \exp(-E/T_1)$ and $R_o \exp(-E/T_2)$ are substituted in equation (4) for $N(T_1)$ and $N(T_2)$. The result is

$$f(T_1)/f(T_2) = \exp(E/T_2 - E/T_1). \tag{5}$$

Equation (5) then is rewritten after substituting $T_2 = T_1 + \Delta T$ as

$$\Delta T = T_1^2 \ln (f(T_1)/f(T_2)/[E - T_1 \ln (f(T_1)/f(T_2))]. \tag{6}$$

Equation (6) is the desired relation between measured fractional releases $f(T_1)$ and $f(T_2)$ and the temperature difference ΔT between T_1 and T_2. It may be of interest to note that only an activation energy but not a frequency factor appears in equation (6).

2. Calculation of Temperatures from Measured Release Rates

(U)   To determine the capability of equation (5) to predict temperatures in the A6 core, release measurements of the 125-day Sn^{123m} isotope on sections of element 1D1G of the A6 core taken at different core stations were carried out. The nuclide Sn^{123m} was chosen as indicator nuclide because of its conveniently long half-l.. and particularly because of the fact that the precursors of Sn in the 123 mass chain have very short half lives so that Sn^{123m} is in fact the diffusing species. The fractional releases of Sn^{123m} from sections of fuel element 1D1G as a function of core station are summarized in Table XXVII.

TABLE XXVII. Release of Sn^{123m} from Element 191G.

Core Station (Inches from Cold End of Element)	Fraction . Sn^{123m} Released During A6 Test
2.25	0.000
7.5	0.000
12.75	0.000
18.0	0.000
23.25	0.008
28.5	0.104
33.75	0.316
39.0	0.581
44.25	0.698
49.5	0.698

(C-RD) Apparently, core station 23.25 was hot enough to release a measurable amount of Sn^{123m} from the fuel element. As fractional release f_1 in equation (6), the release observed at core station 28.5 was used at a temperature of 2020 °K, measured by thermocouple. A value of 2.06×10^4 calories was used as activation energy, which had been deduced from laboratory measurements of the tin release as a function of temperature. This value is also used as an input datum in the FIPDIF code. The element temperatures, predicted by the TNT code[18] and computed from equation (6) are listed in Table XXVIII.

(U) The agreement between predicted and computed fuel element temperatures is very close. These preliminary calculations indicate the feasibility of 'measuring' in-core temperatures which are either difficult or impossible to measure by another means.

(C-RD)

TABLE XXVIII. Comparison between predicted (TNT Code) and computed temperatures of Element 1D1G.

Predicted Temperature (°K)	Computed Temperature (°K)	Predicted-Computed Temperatures T (°K)
2240	2275	-35
2395	2341	+54
2495	2495	0
2510	2495	+15

IX. THEORETICAL CONSIDERATIONS

(U)   The FIPDIF code requires as input data a frequency factor, D_0, and an activation energy, E/R, for each nuclide in its library in order to compute the release of these nuclides by diffusion from an operating NERVA reactor. In order to assign diffusion constants to nuclides whose release has not been measured, a relation between a physical or thermodynamic property of fission product elements whose release had been measured and either the frequency factor D_0 or the activation energy E/R is needed as a guide. The pertinent literature provides very little information as to which physical properties are expected to control the release of fission products by diffusion.

(U)   Faircloth and Flowers[19] observed that metallic fission products which form the least stable carbides escape fastest from a graphite matrix fuel. In more quantitative terms, they postulate that the enthalpy required to evaporate one mole of a fission product from its most carbon-rich carbide is directly related to the diffusion rate of that fission product. On this basis, Faircloth and Flowers suggest in which order fission product nuclides can be arranged according to increasing or decreasing escape rates. Such a sequence of fission product nuclides is not obtained when the nuclides measured at WANL are arranged according to decreasing frequency factors or increasing activation energies.

(U)   Contrary to the observation at WANL, investigators at Battelle[20] believe to have evidence that the values for the diffusion coefficients depend upon the vapor pressures of the fission product elements.

(U)   The thermodynamic and physical properties tested for parallel trends with the measured frequency factors and activation energies were:

    a. enthalpy of vaporization at the boiling point,
    b. vapor pressure at 2000 $^\circ$C,
    c. vapor pressure at the boiling point,
    d. boiling point.

e. atomic radius,

f. ionization potential,

g. electron binding energy,

h. atomic number,

i. thermionic work function, and

j. periodicity in the sense of the Periodic Table of Elements.

(U) No obvious trends between above properties and either D_0 or E/R were formed.

(U) The large variation in frequency factors in Table XVIII, and the lack of any parallel trends between measured frequency factors or activation energies with other physical or thermodynamic properties of fission product elements can be taken as an indication that the fission product release from NERVA fuel cannot be described by a simple diffusion model. Indeed, there is evidence in the pertinent literature[21,22] that surface evaporation should also be included in such a model.

(U) A more complete description of the isothermal steady state release of fission products from NERVA fuel is given by an empirical equation of the form

$$F = (1-K) (f_c + f_S) + K(f_R + f_D) + Q(f_R^1 + f_C^1), \qquad (7)$$

where F is the fraction release, K the fraction of broken beads, f_c the fractional release rate by diffusion through the pyrocarbon coating of the beads, f_S the fractional release rate by sublimation from the surface of the beads, f_R, f_D the fractional release rates from free UC_2 surfaces by recoil and by diffusion, respectively, f_R^1, f_D^1 the fractional release rates due to uranium contamination in the pyrocarbon coating by recoil and by diffusion, respectively, and Q the amount of uranium contamination in the pyrocarbon coating.

(U) Thus far, the derivation of a diffusion constant for a specific nuclide from release rate measurements was based on the assumption

$$F = (1-K) f_c. \qquad (8)$$

The term $Q(f_R^1 + f_D^1)$ in equation (7) is very small because Q was determined to be very small. This term can be neglected. The term $K(f_R + f_D)$ was found to be of the order of 5 percent or less of the total release from 1/4-inch fuel sections. Since the ratio of the number of broken beads versus number of intact beads is appreciably smaller for a full length fuel element than for a 1/4-inch section, the second term can also be neglected in FIPDIF calculations. Above considerations are the justification for using only 'slow' diffusion constants as input data for the FIPDIF code. The good agreement between FIPDIF predictions and the measured release of most mass chains during the various NRX reactor runs confirms the approach for deriving diffusion constants as basically correct. Another implication of these considerations is that the magnitude of f_s compared to f_c must be small. Thus, equation (8) is a reasonable and useful simplification of equation (7).
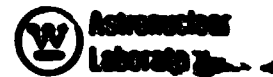
X. POSSIBLE EFFECTS OF NEW FUEL DEVELOPMENTS ON THE SOURCE TERM FOR NERVA REACTORS

(C-RD)   In the following an educated guess is attempted as to the effects of future contemplated changes of NERVA fuel on the fission product release. Such changes are

        a. the use of a higher density graphite matrix than, for example, in A6 fuel,

        b. the replacement of the innermost pyrographite coat around each UC_2 kernel by a spongy modification of graphite,

        c. an additional coat of zirconium carbide around the Triplex beads, and

        d. a thicker pyrocarbon coating around the UC_2 kernel.

(C-RD)   Items a, c, and d are expected to increase the effective diffusion barrier for fission products; their release from R-type reactors at A6 core temperatures most probably will be lower compared with the release from the A6 reactor. However, the higher operating temperature for which the R-type reactors are designed may more than offset the retarding effect of higher diffusion barriers on the fission product release.

(C-RD)   Item b may or may not have any effect on the actual fission product release, since the spongy graphite layer nearest to the UC_2 core is intended to prevent cracking of the pyrographite layers of the bead due to different thermal expansion coefficients of pyrocarbon and UC_2 kernel. Although it is conceivable that the spongy graphite indeed may act as a true sponge by 'soaking up' fission products and thus in effect increase the effective diffusion barrier somewhat more, the existence of such an effect would be rather difficult to verify experimentally.

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